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AIR POLLUTION

APPENDIX E - AIR POLLUTION CASE STUDY OF
THE SOUTH HOLLAND REGION (FIRST PHASE) - 1973,
NUMBER 6

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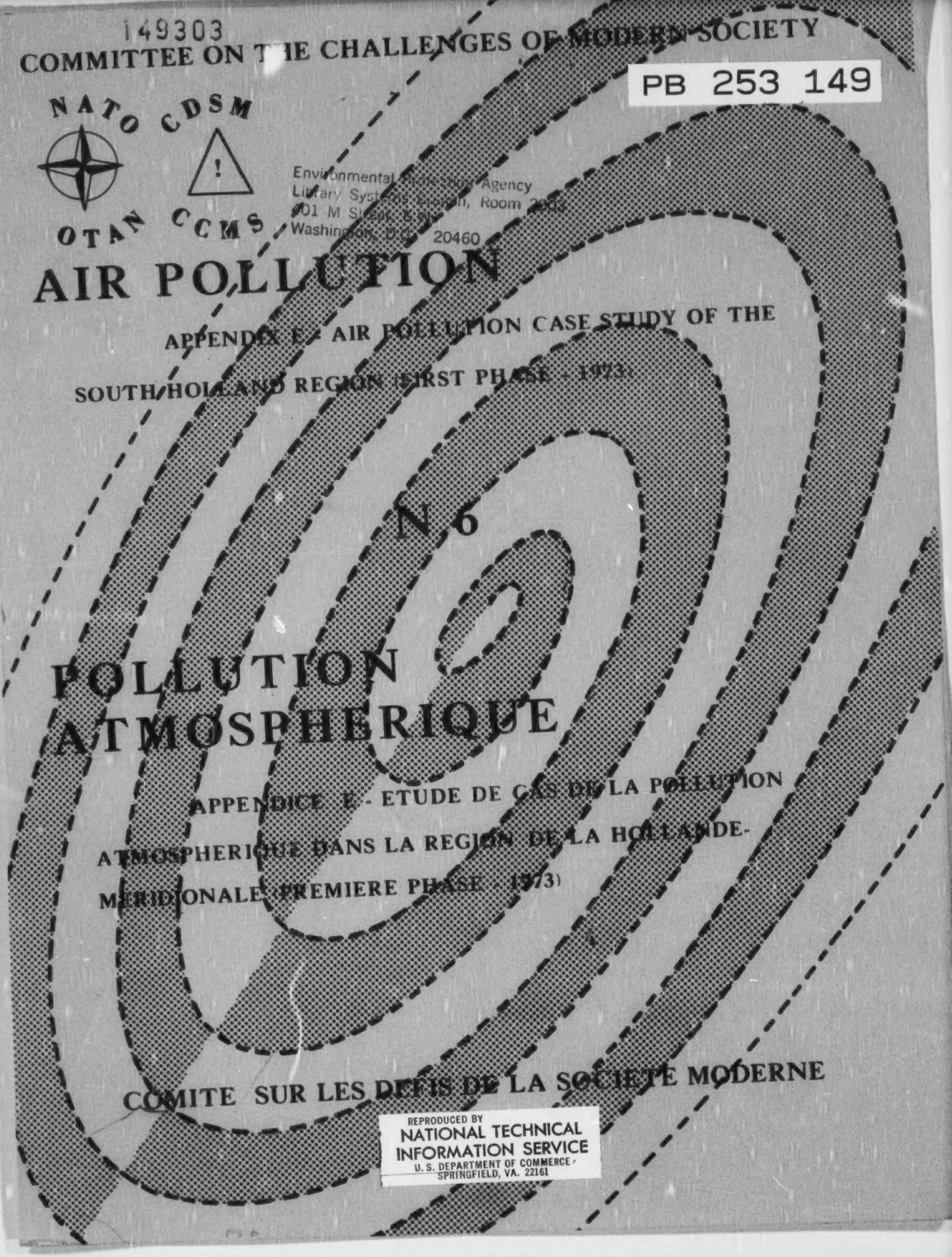

AIR POLLUTION

APPENDIX E - AIR POLLUTION CASE STUDY OF THE SOUTH HOLLAND REGION (FIRST PHASE - 1973)

N 6

POLLUTION ATMOSPHERIQUE

APPENDICE E - ETUDE DE CAS DE LA POLLUTION ATMOSPHERIQUE DANS LA REGION DE LA HOLLANDE-MERIDIONALE (PREMIERE PHASE - 1973)

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This report summarizes an experimental investigation of air pollution in the South Holland Region to analyze the air with respect to paramenters that might affect the air pollution in South Holland. Emphasized are the differences in air pollution during periods with and without photochemical smog development. The investigation was meant tolook intothe occurence and possible prevention of combinations of air pollution parameters. The Institute for Plant Disease Investigations in Wageningen took on the additional task of investigating the effect of air pollution on the development and growth of various plants with the aid of test plots and test greenhouses. The results of this investigation in 1973 areadded to this report as appendices IIIA and IIIB.

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AIR POLLUTION CASE STUDY OF THE SOUTH HOLLAND REGION

FIRST PHASE (1973)

by

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1. Introduction

The purpose of the experimental investigation of air pollution in the South Holland Region is to analyze the air with respect to parameters that possibly might affect the air pollution in South Holland. Emphasized are the differences in air pollution during periods with and without photo chemical smog development. Early in 1972 a planning session met to initiate an efficient investigation. Participants were the National Institute of Public Health (R.I.V.), air pollution experts of South Holland, the National Meteorological Institute (K.N.M.I.) and the Ecology Department of the Inspectorate of Public Health. After approval of the plan was obtained, its execution was started in the middle of 1972. At the end of 1972 the first field measuring stations and several measuring instruments could be installed and tested. At the end of 1972 the first cars equipped with instruments, which are referred to as "mobile units," were ready, so in the beginning of 1973 the first mobile measurements could be made.

To the chemical-physical investigation with the aid of stationary and mobile stations, another project was added. The Institute for Plant Disease Investigations (I.P.O.) in Wageningen investigated the effect of air pollution on the development and growth of various plants with the aid of test plots and test greenhouses. The results of this investigation in 1973 are added to this report as appendices IIIA and IIIB.

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The experimental investigation is principally meant to look into the occurrence and possible prevention of combinations of air pollution parameters. However, the obtained data can also be used by the regional guarding and emergency authorities during periods of high air pollution.

2. The Mobile Units

During periods of very high air pollution, two of the mobile units continually provided data from fixed locations (I.G./T.N.O.: at Hoek van Holland, Monster and Maassluis and the R.I.V. mobile units at Schiedam). The mobile units at The Hague and Rotterdam were available for checking up on incoming complaints.

During periods of high air pollution the four cars were used separately. The sampling scheme for each of the cars was different in order to find out after a year which scheme would be most efficient.

The K.v.W. car was used in Rotterdam. Its scheme was to obtain measurements for a half hour at each of eighteen locations in different parts of the city. In this way three days of measurements during a one week period provided the complete sampling pattern. Appendix IV contains the data obtained by this car. The interpretation of the collected data made it possible to decide in what way the car could be used most efficiently. The car in The Hague obtained samples in The Hague region and the suburbs of Rijswijk, Voorburg, Leidschendam and Zoetermeer. At six locations measurements were made, one day at each location. The car of I.G./T.N.O. sampled at three locations, with one week of continual measurements at each location.

The cars measured NO_2, NO, O_3 and CO. The data were registered by a datalogger on a telex with paper tape.

3. The Stationary Measuring Stations for the Experimental Investigation

The stationary measuring stations are located at Vlaardingen (20 Floreslaan), Naaldwijk (test center for vegetables and fruits, 38 Zuidweg), Rotterdam-C (K.v.W., 74 Baan), The Hague (B.W.T., 9011 Mauritskade) and Hellevoetssluis (Oude Werf, Rijkswaterstaat).

The initial half period monitors were installed for NO, NO_2 and ozone at the fixed stations. A few stations (Vlaardingen, The Hague and Rotterdam) also measured hydrocarbons and dust. Beginning in September, 1973, "on-line" data from the mentioned stationary stations were obtained at Bilthoven.

No reference standard (i.e., a verification gas system) was available for the precise testing of the used instruments in 1973. This severely restricted the possibilities in interpreting the data.[1] However, some usable measurement series for various components were already available in 1973. To investigate possible relations between sulphuric acid aerosol and the many complaints during extended periods of high air pollution, sulphuric acid was measured in the Rijnmond region, too. Results are given in appendix I.

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[1] This reference standard was installed at the R.I.V. and put to use during 1974.

Some research is done on the data obtained in November and December, 1973, and in March, 1974. The results can be found in appendix II. More elaborate data processing techniques are emphasized. Superficially seen, the transparency of the data diminishes after these procedures are performed on it, but this is considered unavoidable in a rather rigorous evaluation of the available data.

4. Conclusions

Based on the results of processing a part of the measured data we can conclude the following:

(1) At Vlaardingen for most polluting components, especially acetylene (C_2H_2), the highest concentrations were measured in the morning with unstable atmospheric conditions.[1]

(2) Winds from the north cause higher acetylene (C_2H_2) concentrations originating from traffic emissions.

(3) The polluting components nitrogen dioxide (NO_2), iso butane ($i.C_4H_{10}$), butane ($n.C_4H_{10}$) and propane (C_3H_6) have maximal average concentrations in Vlaardingen with southwest winds. This indicates that these components originate at some distance. Higher concentrations of nitrogen monoxide (NO) and ethene (C_2H_4) occur at very low wind velocities.

(4) The ratio of nitrogen monoxide to nitrogen dioxide ($NO : NO_2$) diminishes at Vlaardingen with higher wind velocities for

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[1] In the period October 1973 - March 1974, now and then very high concentrations of hydrocarbons were measured, especially for the components acetylene, butane and propene (see appendix II).

all directions except south-southeast. For higher wind velocities from this direction a relatively high NO concentration is measured. This might come from a NO_x source at some distance from the station.

(5) The highest concentrations of NO are measured at locations adjacent to highways.

(6) The data from the mobile units (cars) showed that observed ground concentrations of NO and NO_2 in the region Rotterdam-Schiedam are coming 25-30% of the time from sources in the direction of the Rijnmond industrial region. At Vlaardingen this contribution was assessed to be circa 50%.

(7) The measured sulphuric acid concentrations are low. The average value was approximately $0.3\,\mu g/m^3$; maximal values of approximately $1\,\mu g/m^3$ were measured during periods of relatively high SO_2 levels.

(8) The tests with artificial PAN gas applications to plants showed that the lettuce varieties grown in the Westland region are not very sensitive to PAN. The small stinging nettle, used as an indicator plant, is very sensitive; street grass is less sensitive. The gas treatment experiment with ozone showed the extremely high sensitivity of the tobacco variety Bel W3.

(9) At various stages during the growing season of the indicator plants damage to the plants was observed. It was caused by ozone (for tobacco) and peroxyactylnitrate (for small stinging nettle and street grass); for hydrogen fluoride a gradual development of the damage during the growing season was observed (gladiolus and freesia). In

the small greenhouses that were supplied with an active carbon filter for removal of the air pollution components, the plant growth was better than the growth of plants in greenhouses without a filter.

(10) It appeared to be advantageous to display the measured data of multiple air pollution components, measured at one location, for example at Vlaardingen, in so-called score diagrams. The position of an hour-measurement in such a diagram can give an insight into the relation between the hour-measurement and other factors, for example, atmospheric stability. This position is calculated from a data file previously classified with respect to the hour-measurement factor. The hour-measurement is not necessarily a part of this data file.

APPENDIX I

Sulphuric Acid in the Atmosphere

Sulphuric acid is found in the atmosphere in the form of particles. It is sampled with the aid of inert filters. From the aerosol sample the sulphuric acid is selectively extracted from the sulphate salts. In the extract first the amount of sulphate representative for the sulphuric acid on the filter is measured. The selectivity is so high that in all cases no increase of the measured sulphuric acid caused by the presence of sulphates can be shown. This combination of selective extraction of sulphuric acid and the specific measurement of sulphate certifies a selective measurement of the sulphuric acid on the filter. No other method in use does this.

The greatest problem, not yet solved, is the question: how well does the sample represent the concentration in the atmosphere?

The amount of sulphuric acid on the filter will be representative only when the following conditions are met:

(a) all sulphuric acid is caught by the filter;

(b) no sulphuric acid is formed on the filter;

(c) sulphuric acid is not transformed into something else.

The first condition is not difficult to meet because of the size of the sulphuric acid particles. The other conditions are more bothersome. Formation of sulphuric acid is not impossible when the sampled material contains components that work as a catalyst for the SO_2, for example, Fe^{2+}, Fe^{3+}, Mn^{2+}, Co^{2+}, Ni^{2+}.

The conversion of the sulphuric acid is not impossible either when basic components (for example, NH_3) are present in the air.

The magnitude of both effects increases with the sampling time. Short sampling times were impossible not long ago, because of the large sample sizes that were required.[1]

Average day concentrations are shown in Tables I-III. Average concentrations are for:

Vlaardingen:	0.26 $\mu g/m^3$
Rotterdam:	0.21 $\mu g/m^3$
Ommoord:	0.20 $\mu g/m^3$

Concentrations below the detection limit (0.2 $\mu g/m^3$) are reported as 0.1/m^3. High sulphuric acid concentrations are observed before and after periods of higher sulphur dioxide concentrations.

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[1] It is possible to improve the analyzing methods by using very sensitive isotope dilution techniques. Sampling with an interval time of ten minutes becomes possible in this way.

Table I

H_2SO_4 and SO_2 in the Atmosphere in µg/m^3 at Rotterdam for 1973

Day Dag	September		Oktober		November		December	
	H_2SO_4	SO_2	H_2SO_4	SO_2	H_2SO_4	SO_2	H_2SO_4	SO_2
1	-		0.1	30	0.1	116	0.7	146
2	-		0.1	41	0.1	146	0.7	-
3	-		0.1	26	0.1	105	0.1	-
4	-		0.1	143	0.1	38	0.1	-
5	-		0.4	131	0.1	45	0.1	-
6	-		0.3	143	0.1	64	0.3	-
7	-		0.3	68	0.1	113	0.1	71
8	-		0.1	75	0.1	154	0.1	53
9	-		0.1	79	0.1	64	0.1	49
10	-		0.1	64	0.1	26	0.1	-
11	-		0.3	45	0.1	30	0.1	131
12	-		0.3	109	0.1	113	0.1	161
13	-		0.4	94	0.1	90	0.1	94
14	-		0.5	124	0.1	56	0.1	4
15	-		0.1	143	0.1	53	0.1	23
16	-		0.1	105	0.1	53	0.3	41
17	-		0.1	79	0.1	34	0.3	60
18	-		0.1	-	0.1	131	0.1	105
19	-		0.1	86	0.4	53	0.3	-
20	-		0.1	79	0.1	41	0.4	83
21	-		0.1	41	0.1	281	0.4	120
22	-		0.1	120	0.4	225	-	
23	-		0.1	45	0.4	124	-	
24	0.1	83	0.1	150	0.3	90	-	
25	0.1	60	0.1	-	0.4	15	-	
26	0.1	135	0.1	203	0.3	8	-	
27	0.1	53	0.6	229	0.6	11	-	
28	0.1	56	0.1	-	0.7	83	-	
29	0.1	41	0.1	94	0.7	30	-	
30	0.1	38	0.1	64	0.5	68	-	
31	-		0.1	68	-		-	

0.1 means < 0.2

Table II

Sulphuric Acid in the Atmosphere in $\mu g/m^3$ at Vlaardingen

Day Dag	September	Oktober	November	December
1	-	0.1	0.4	0.7
2	-	-	0.1	0.7
3	-	-	0.1	0.5
4	-	-	0.1	-
5	-	-	0.1	0.1
6	-	0.1	0.1	0.1
7		0.1	0.1	0.4
8	-	0.1	0.1	0.1
9	-	0.1	0.1	0.1
10	-	0.1	0.3	-
11	-	0.1	-	-
12	-	0.1	0.4	0.1
13	-	0.3	0.4	0.1
14	-	0.1	0.6	0.1
15	-	0.4	0.1	0.1
16	-	0.6	0.5	0.1
17	-	0.3	0.5	0.1
18	-	0.1	0.1	0.3
19	0.1	0.1	0.5	1.1
20	0.1	0.1	0.6	-
21	-	0.1	0.7	-
22	0.6	0.1	0.5	-
23	0.1	0.1	0.4	-
24	0.1	0.1	0.1	-
25	0.1	0.1	0.1	-
26	0.1	0.1	0.7	-
27	0.1	0.5	0.5	-
28	0.1	0.1	0.7	-
29	0.1	0.1	0.6	-
30	0.1	0.1	0.6	-
31	-	0.1	-	-

0.1 means < 0.2

Table III

Sulphuric Acid in the Atmosphere in $\mu g/m^3$ at Ommoord

Day Dag	September	Oktober	November	December
1	-	0.1	0.1	0.6
2	-	0.1	0.2	0.6
3	-	0.1	0.7	0.1
4	-	0.1	0.1	0.1
5	-	0.1	0.1	0.1
6	-	0.1	0.1	0.1
7	-	0.1	0.1	0.1
8	-	0.1	0.1	0.1
9	-	0.1	0.1	0.1
10	-	0.1	0.1	0.4
11	-	0.1	0.1	0.3
12	-	0.1	0.1	0.4
13	-	0.1	0.3	0.1
14	-	0.1	0.3	0.1
15	-	0.1	0.3	0.1
16	-	0.1	0.3	0.1
17	0.1	0.1	0.4	0.3
18	0.1	0.1	0.1	0.1
19	0.1	0.1	0.3	0.3
20	0.1	0.1	0.4	0.4
21	0.1	0.1	0.1	0.4
22	0.1	0.1	0.5	-
23	0.1	0.1	0.3	-
24	0.1	0.1	0.1	-
25	0.1	0.1	0.1	-
26	0.1	0.1	0.7	-
27	0.1	0.5	0.5	-
28	0.1	0.1	0.7	-
29	0.1	0.1	0.6	-
30	0.1	0.1	0.6	-
31	-	0.1	-	-

0.1 means < 0.2

APPENDIX II

Experimental Processing of the Stationary Measuring Stations

1. Introduction

A part of the current available data, obtained by the project Air Pollution South Holland, is used for some experimental processing.

The question is: what processing should be used to understand the parameters related to photo chemical smog? A short overview of the data obtained from the stationary measuring stations is given. Special attention is given to the Vlaardingen station, which provided average hour concentrations of 10 air pollution components.

An attempt is made to define the most important relations between the components for various atmospheric conditions. In addition an attempt is made to find a clear presentation that would show the combined measurement results.

2. Available Data

For the project Air Pollution South Holland, a number of stationary stations measured the following air pollution components:

Ozone	O_3	Ethene	C_2H_4	ISO-butane	$i\text{-}C_4H_{10}$
Nitrogen dioxide	NO_2	Ethane	C_2H_6	Butane	$n\text{-}C_4H_{10}$
Nitrogen monoxide	NO	Propene	C_3H_6		
Acetylene	C_2H_2	Propane	C_3H_8		

Note: The figures with roman numerals are included at the end of the appendix; the remaining figures and tables are included in the text of the appendix itself.

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O_3, NO_2 and NO are measured at the stations with ENRAF-NONIUS monitors. The concentrations are measured colorometric as half-hour averages. The measured O_3 concentrations were for technical reasons not usable for processing. The concentrations of hydrocarbons were measured at the stationary stations at Vlaardingen and Rotterdam-C with the aid of an Askol gas chromotograph.

The data from the stations were sent to the National Institute for Public Health at Bilthoven.

The locations of the stations are shown in Fig. I according to the code given in Table 1.

In addition the locations of some semi-automatic NO_2 measuring stations are given;[1] these data are compared with the results of the ENRAF-NONIUS instruments.

Hour averages obtained in November 1973, December 1973, and March 1974, are processed first because only a low percentage of data is missing for these months.

For the study of the relation between the various components, only data obtained at hours for which measurements of all components are available can be used. For this reason only data from December 1973, and March 1974, are used in the analysis of data from station Vlaardingen.

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[1] Six-hour averages measured with the colorometric Saltzman-method.

The National Meteorologic Institute provided information about meteorologic parameters: direction of the wind, wind velocity and the atmospheric stability, measured at Zestienhoven.

Table 1

Stationary Automatic Measuring Stations

VI	The Hague
V2	Rotterdam-C
V3	Vlaardingen
V4	Naaldwijk
V5	Hellevoetsluis
V6	Rotterdam-Ommoord

Semi-automatic NO_2 Measuring Stations

H1	The Hague
H5	Naaldwijk
H9	Vlaardingen

3. Processing

3.1. Comparison of the Data of Automatic (Stationary) and Semi-Automatic Measuring Stations

The monthly averages of NO_2 concentrations of the automatic stations (the Hague-V1, Naaldwijk-V4 and Vlaardingen-V3) are compared with the concentrations of the semi-automatic stations, H1, H5 and H9, respectively (Table 2).

Table 2

Monthly Averages of NO_2 Concentrations in ug/m^3

H Semi-automatic and V Automatic

	Den Haag		Naaldwijk		Vlaardingen	
November	H1	40	H5	36	H9	47
	V1	44	V4	40	V3	47
December	H1	43	H5	38	H9	46
	V1	41	V4	42	V3	21

The monthly average concentrations are pretty much the same for both months. Only at Vlaardingen for December does there occur a great difference in monthly average concentration.

3.2. Analysis of Data from the Vlaardingen Station

The hour average concentrations at Vlaardingen (V3) were processed. Considered were (a) the air pollution components and their mutual relationships, and (b) the meteorological parameters: direction of the wind, wind velocity and atmospheric stability. The measured concentrations and the computed relations were classified according to direction of wind, wind velocity and stability. For a closer study of the relation between the measured concentrations and the atmospheric stability a discriminant analysis is made.

3.2.1. Classification According to Direction of Wind

The hourly observations of the direction of the wind at Rotterdam (Zestienhoven) were used to make a partition in eight sectors. Sector 1 contains directions of wind in between 0° and 45° (NNE). The average concentrations for each wind direction sector and component are shown graphically in Figs. II through VI. The length of the bars corresponds to the average concentration; the scale ($\mu g/m^3$) is indicated with concentric circles.

The average wind velocities for each sector are shown in Fig. VII.

The number of hourly observations N, from which the averages of a given sector are calculated, are shown in Table 3 for the months of December 1973, and March 1974.

Table 3

Number of (Hourly) Observations for Each Wind Direction Sector

Wind Direction	NNE	ENE	ESE	SSE	SSW	WSW	WNW	NNW
Sector	1	2	3	4	5	6	7	8
Number of Hours:								
December 1973	28	4	25	54	169	111	92	53
March 1974	66	155	61	62	70	107	34	46

The levels of the measured concentrations differ widely because of the differences in the weather. These differences in the weather between December and March show markedly in the frequencies of the observed wind directions (Table 3). Both December and March measurements for wind directions in Sector 1 show higher concentrations for C_2H_2 and C_3H_6. Especially for December, and also for wind directions in Sector 4 (SE), high concentrations of C_2H_4, C_2H_6, C_3H_8, i-C_4H_{10} and n-C_4H_{10} are found.

The higher concentrations for C_2H_2 might be caused by traffic emissions because acetylene is emitted in particular by traffic. Moreover, no major industrial sources are located northeast from the test station at Vlaardingen.

3.2.2. Classification According to Wind Velocity

The December concentration measurements with SSW wind (Sector 5) are classified according to wind velocity.

The hourly observations were divided into seven groups according to the average wind velocity of that hour. The width of the class is 4 knots (2 m/s); the first one contains wind velocities from 0-4 knots; the seventh group (> 24 knots : 1 observation) is not used. The average concentrations ($\mu g/m^3$) in each wind velocity group are shown in Figs. VIII and IX.

Table 4 shows the number of observations for each wind velocity group.

Table 4

Number of Hours for Each Wind Velocity Group
for Wind Direction Sector 5
(December 1973, Vlaardingen)

Wind Velocity Group	Sector 5, SSW Number of Hour Samples per Group
0 - 4 knots	7
5 - 8 knots	24
6 - 12 knots	48
13 - 16 knots	69
17 - 20 knots	10
21 - 24 knots	10
	168

For the components NO_2, i-C_4H_{10}, n-C_4H_{10} and C_3H_6 the maximal average concentrations occur for higher wind velocities. This indicates that these components arrive from some distance.

For NO and C_2H_4 higher concentrations occur at very low wind velocities (0-4 knots or 0-2 meters/sec). The average concentrations per wind velocity class calculated from the data of March show the same trend.

Figure X shows the ratio NO:NO_2 as a function of the wind velocity for each wind direction sector. This ratio decreases with increasing wind velocity; this is the case for all wind directions except for the wind direction sector 4 (SSE). For higher wind velocities a relatively high NO concentration for this sector is measured. Probably this originates from industrial sources at some distance from the station.

3.2.3. Classification According to Stability Class

The total number of hour samples (December: 554; March: 601) is also classified according to atmospheric stability.

Six stability classes, A, B, C, D, E and F, as proposed by Pasquill, are used. The classes are based on wind velocity at a 10-meters height, radiation and cloudiness. The K.N.M.I. Meteorologic Institute provided a classification based on cloudiness and wind velocity; radiation information was not (yet) available.

Meaning of the symbols:

A	very unstable
B	medium unstable
C	slightly unstable
D	neutral
E	slightly stable
F	medium through very stable

The number of samples per class are shown in Table 5.

Table 5
Number of Hour Samples per Stability Class

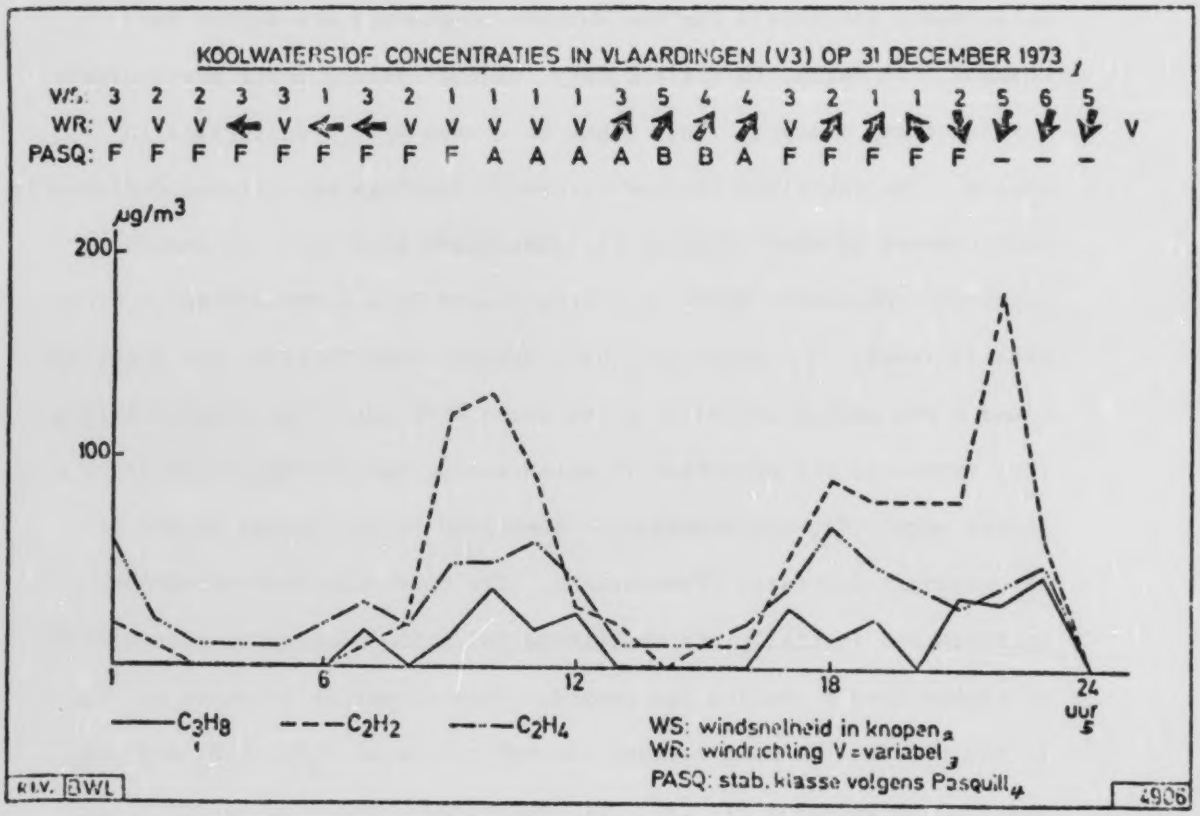
Stability Class	A	B	C	D	E	F
Number of Hour Samples:						
December	5	9	12	479	20	29
March	5	30	20	409	28	72

The average concentrations per component and per stability class for December 1973, and March 1974, are shown in the Figs. XI - XIV. Higher concentrations occur, particularly in the classes A, B, E and F. In December the levels for the various components are higher for class A. In March, on the contrary, maximal values occur particularly for stability class B. This might be caused by the difference in season. The stability classes for March (spring) are defined differently with respect to wind velocity and cloudiness than those of December (winter). Moreover, March is characterized by a lower average concentration level. The relatively high average contributions for stability class A are mainly caused by a few hours with very high concentrations. This refers to the situation in which during the morning hours after a stable night (F), the atmospheric stability in the lowest layers of air strongly decreases (fumigation). The convective air movements increase,and initially air containing polluting components accumulated at higher levels reaches the ground. This situation is shown in Fig. 1. In addition to the hour average concentrations of C_3H_8, C_2H_2 and C_2H_4 measured on December 31, 1973, at Vlaardingen, the wind velocity and stability are given here.

The stability is classified F until 10:00 a.m.; afterwards it is A. The K.N.M.I. Meteorological Institute's system does not include class A at 10:00 a.m. in the winter because a difference is made for day and night in the definition of the stability classes. However, it is likely because of the observed increase of the hydrocarbon concentrations that the stability decreases already at 8 a.m.

Fig. 1

Hydrogen Carbon Concentrations at Vlaardingen (V3)
December 31, 1973



Key:

1. Hydrogen carbon concentrations at Vlaardingen (V3), December 31, 1973
2. WS: wind velocity in knots
3. WR: wind direction V = variable
4. PASQ: stability class according to Pasquill
5. Time

The above shows that for most components the highest concentrations are found in the morning for the stability classes A and B.

In order to get some understanding of the absolute levels and the mutual relationships of the air polluting components, in particular with respect to the atmospheric (clearance) conditions, the concentrations of the 10 measured components are shown in Figs. XI -XIV in histograms, classified according to stability.

However, it is impossible in this way to show the relation between the concentration levels as a function of the stability, from hour to hour.

Therefore, a multiple discriminant analysis is given.

4. Discriminant Analysis

4.1. General

The purposes of the discriminant analysis were:

(a) To find the significance of the classification of the various air polluting components measured at Vlaardingen according to stability as defined by Pasquill;

(b) To look for differences in composition of the totality of all polluting components for the various stability classes;

(c) To describe the daily course of the concentration levels for multiple components in a simple diagram that would show the relation with the stability classes.

The multiple discriminant analysis is a form of multiple regression analysis which treats the classification problem as a regression problem.

Observations of a number of variables are classified in two or more groups according to a previously established set of criteria. Now a set of so-called discriminant functions is derived from the form:

$$d + a_1x_1 + a_2x_2 \ldots\ldots\ldots\ldots\ldots\ldots a_px_p$$

with a_1, $a_2 \ldots\ldots\ldots a_p$ as discriminant coefficients. These are calculated in such a way that the discriminant scores d, for the various measured values of the explanatory variables x_1, x_2, $\ldots\ldots x_p$, differ as much as possible with respect to each other.

The coefficients a_1, $a_2 \ldots\ldots a_p$ for the first discriminant function are the elements of the vector a_I, that can be calculated from:

$$(B - \lambda_I W)\ a_I = 0$$

with B: matrix of the covariances between the two groups

W: matrix of the covariances within the groups

λ_I: eigenvalue corresponding to the (eigen) vector a_I.

The vector a_I is calculated as the first vector of the non-symmetric matrix $W^{-1}B$.

The coefficients of the second discriminant function d_{II} are found as elements of the second eigenvector of the matrix $W^{-1}B$; the corresponding eigenvalue is λ_{II}.

The corresponding eigenvalues indicate the discriminating ability of the discriminant functions. The method described by Hope (1) is used here.

4.2. Computations

The analysis is performed for nine variables, that is, nine air polluting components. The data from March 1974, are classified according to three groups:

Group	Hour Average for Observed Stability Classes
AB	A or B
CD	C or D
EF	E or F

Each hour observation, X_1, X_2......X_9, consists of nine hour values of the components NO_2, NO, C_2H_6, C_2H_4, C_3H_8, C_2H_2, i-C_4H_{10}, n-C_4H_{10} and C_3H_6.

For the assessment of the discriminant coefficients only part of the available data are used.

Inspection of the available data indicates an occurrence of lower concentrations of hydrocarbons for lower NO:NO_2 ratios. Lower ratios are observed, especially in class D, the most frequently occuring class (March: 409 hours).

Lower concentration levels are considered to be less interesting at this stage; therefore only hour observations for which

$$\frac{(NO)}{(NO_2)} > 1$$

are included in the groups.

The groups contain the following number of observations:

Group AB: 10
CD: 58
EF: 28.

The explanatory variables are considered to have a normal distribution. Therefore, on the base of the assumed log-normal distribution of the components, the logarithms of the component concentrations are used for variables. The coefficients of the discriminant functions

$$d_I = a_{I,1} \log NO_2 + a_{I,2} \log NO + \ldots. + \ldots. + a_{I,9} \log C_3H_6$$

and

$$d_{II} = a_{II,1} \log NO_2 + a_{II,2} \log NO + \ldots + a_{II,9} \log C_3H_6$$

are calculated as follows:

	I	II
NO_2	$a_1 = -0.50$	$a_1 = 0.27$
NO	$a_2 = 0.53$	$a_2 = -0.14$
C_2H_6	$a_3 = -0.49$	a_3 -
C_2H_4	a_4 -	a_4 -
C_3H_8	$a_5 = 0.28$	a_5 -
C_2H_2	$a_6 = 0.39$	$a_6 = 0.14$
$i\text{-}C_4H_{10}$	a_7 -	$a_7 = -0.54$
$n\text{-}C_4H_{10}$	a_8 -	$a_8 = 0.66$
C_3H_6	a_9 -	$a_9 = -0.38$
	$\lambda_I = 0.61$	$\lambda_{II} = 0.20$

The coefficients with value "-" are smaller than 0.10 and are not used in the calculations.

The coefficients are such that the group discriminating ability of both functions d_I and d_{II} is maximal. For d_I ($\lambda_I = 0.61$) this is about three times higher than for d_{II} ($\lambda_{II} = 0.20$).

In the next step the discriminant scores, that is, the function values d_I and d_{II}, for each sample (hour) of the groups AB, CD and EF are computed. These scores can be displayed in a so-called score diagram with d_I along the x-axis and d_{II} along the y-axis.

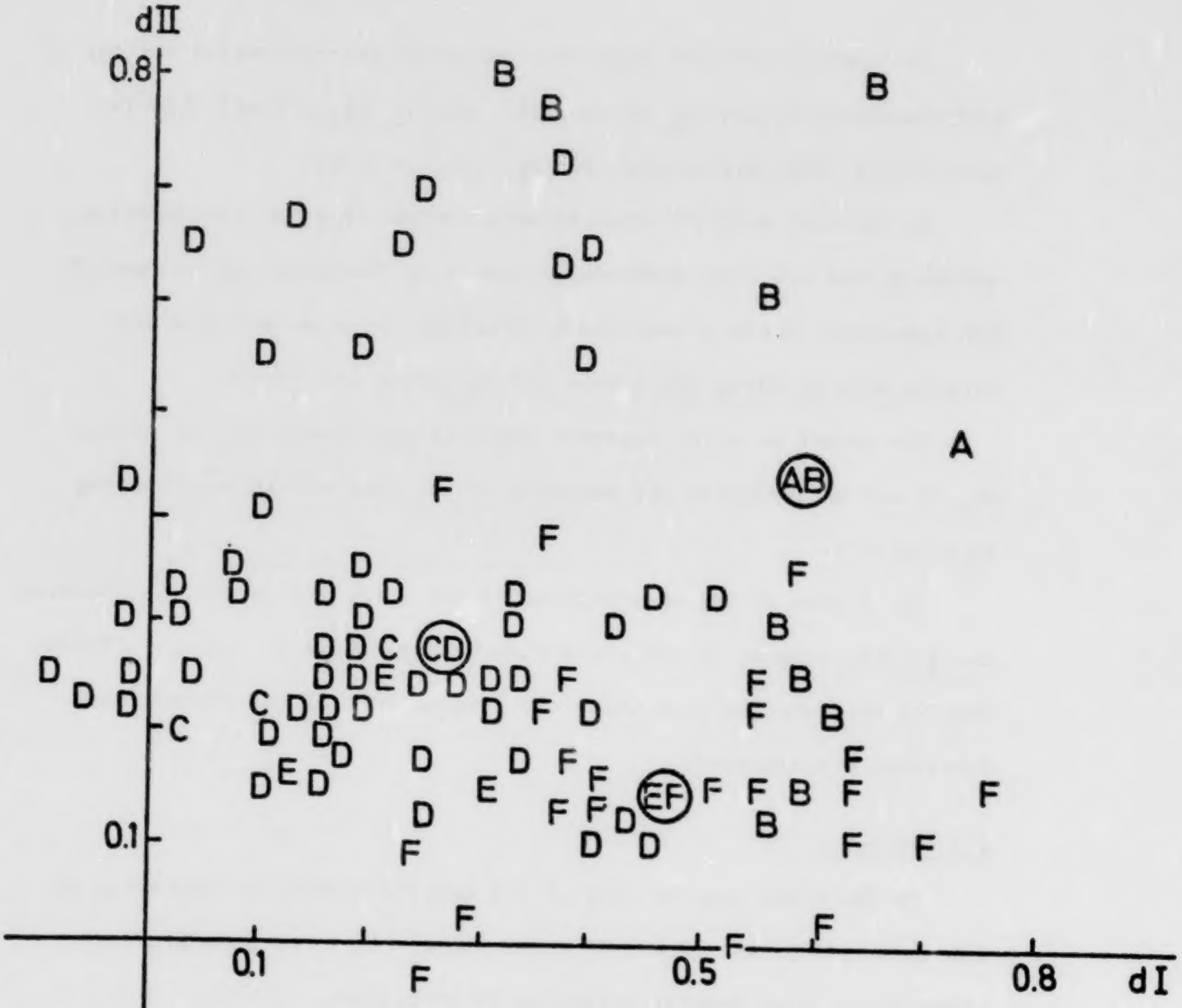
The extent to which separate clusters are formed for the groups AB, CD and EF indicates the adequacy of the classification according to stability.

The scores of the observations of the three groups and, in addition, the gravity centers of the three clusters are shown in Fig. 2. These are the observations from which the coefficients of the discriminant functions are assessed.

4.3. Results

In the score diagram (Fig. 2) a certain degree of clustering of the three groups AB, CD and EF takes place. It seems possible to classify the hour samples according to stability.

The CD centroid has a relatively low score for d_I; this means that for the stability classes C and D the concentrations of NO_2 and C_2H_6 are high; for NO, C_3H_8 and C_2H_2, relatively low with respect to the corresponding concentrations of the classes AB and EF. This is because higher values for NO_2 and C_2H_6 and lower values for NO, C_3H_8 and C_2H_2 will, according to the discriminant function d_I, effect a lower discriminant score (function value).



Score diagram voor de uurwaarnemingen uit de groepen AB, CD, EF

Ⓐⓑ = centroid groep AB
(pasq. klassen A en B)

FIGURE 2 : Score Diagram for the Hour Samples of the Groups AB, CD, EF

Ⓐⓑ = centroid group AB
(Pasquill classes A and B)

The score differences for EF and AB on the d_I axis are small. The d_{II} axis shows a contrast of NO_2, C_2H_2, n-C_4H_{10} (positive coefficients) with NO, i-C_4H_{10}, C_3H_6 (negative coefficients).

So the ratio NO:NO_2 will be lower for the category AB than for EF. In general, hour samples with a low NO:NO_2 ratio will shift in the direction of the fourth quadrant (high d_{II} value, low d_I value).

The score diagrams are drawn for two days of March. From the concentrations of each hour the scores were computed according to the functions d_I and d_{II}. The stability class observed for each hour is given with each score. An underscore indicates that this hour sample is not part of one of the three groups (AB, CD, EF) from which the coefficients are assessed.

(a) March 3, 12 p.m. - March 4, 6 a.m.

The discriminant scores for this period are, together with the computed centroids of the groups AB, CD and EF, shown in Fig. 3.

From 12 p.m. - 5 p.m. the concentration levels are low; at 2 p.m. for i-C_4H_{10}, n-C_4H_{10} and C_3H_6, concentrations of $\pm$ 100 $\mu g/m^3$ are measured. No observations for 6 and 7 p.m. are available. Starting at 8 p.m. higher concentrations are measured for stability class F. At 11 p.m. the measured values for the hydrocarbons are extremely low; the score for this hour is located close to the EF centroid, mostly caused by an NO:NO_2 ratio that stabilizes at a high level. At midnight extremely high concentrations are measured (Table 6). The hour sample taken at 12 a.m. is intentionally omitted from the groups AB, CD and EF, from which the coefficients are computed, although NO:$NO_2 > 1$ for this sample.

Fig. 3: Score Diagram for March 3, noon, through March 4, 6 a.m., 1974

Score diagram voor 3maart 12uur tot 4maart 6uur (1974)

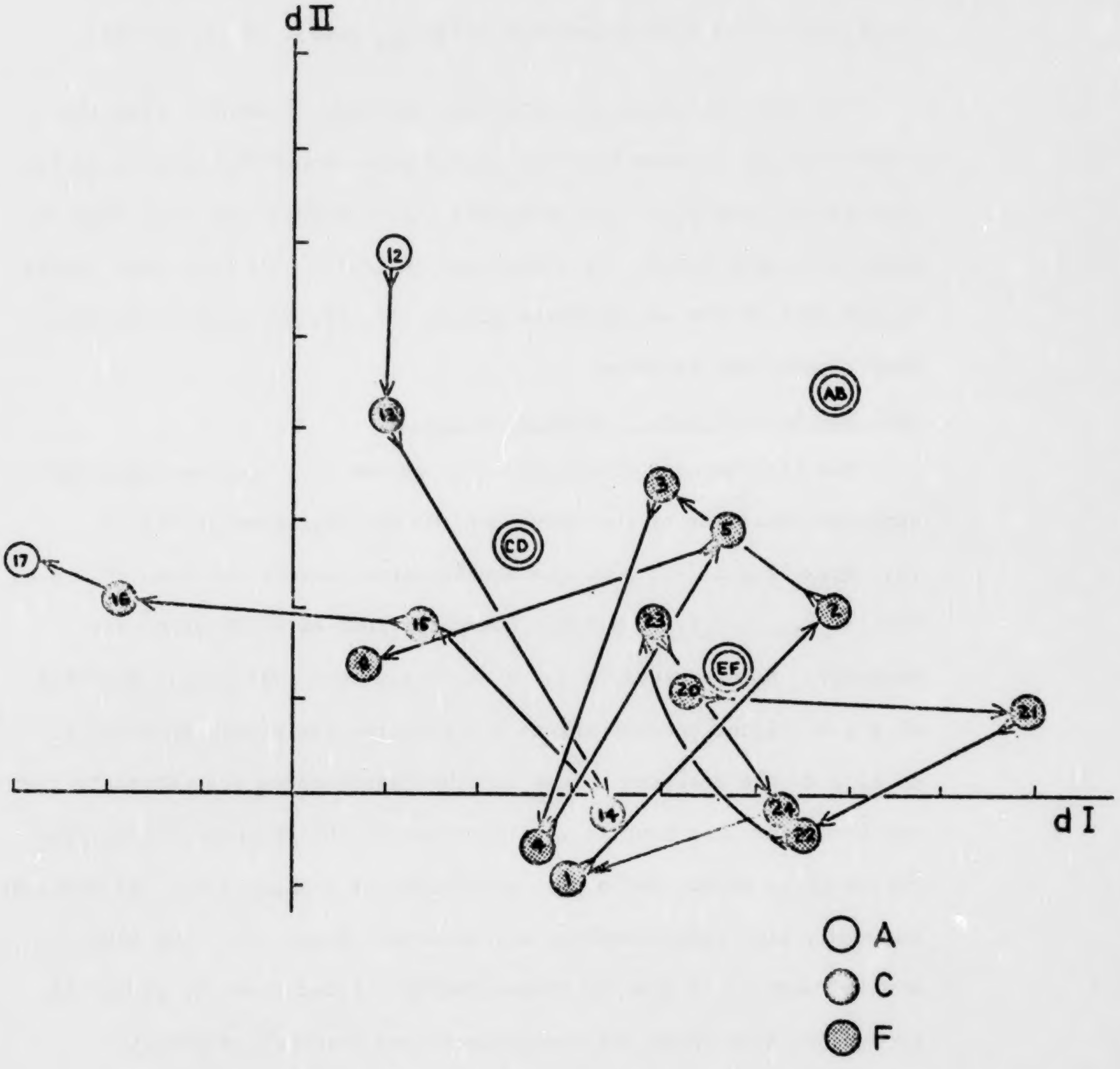


Table 6

Hour Averages of Concentrations in $\mu g/m^3$ Measured at 12 p.m., March 3, 1974, at Vlaardingen

NO_2 : 78
NO : 290
C_2H_6: 45
C_2H_4: 0
C_3H_8: 105

C_2H_2 : 280
i-C_4H_{10} : 560
n-C_4H_{10} : 930
C_3H_6 : 960

Stability Class F

Wind Direction 40°
(Sector 1)

Wind Velocity 2 knots

The discriminant scores for this hour (d_I and d_{II}) agree with the scores computed for 10 p.m. However, at 10 p.m. no hydrocarbon concentrations > 100 $\mu g/m^3$ are measured. Based on this analysis, the hour sample from 12 a.m. cannot be considered to be a deviation. It will be checked whether this sample is correctly measured.

The following hour samples from March 4, 1-6 a.m. do not show any special features.

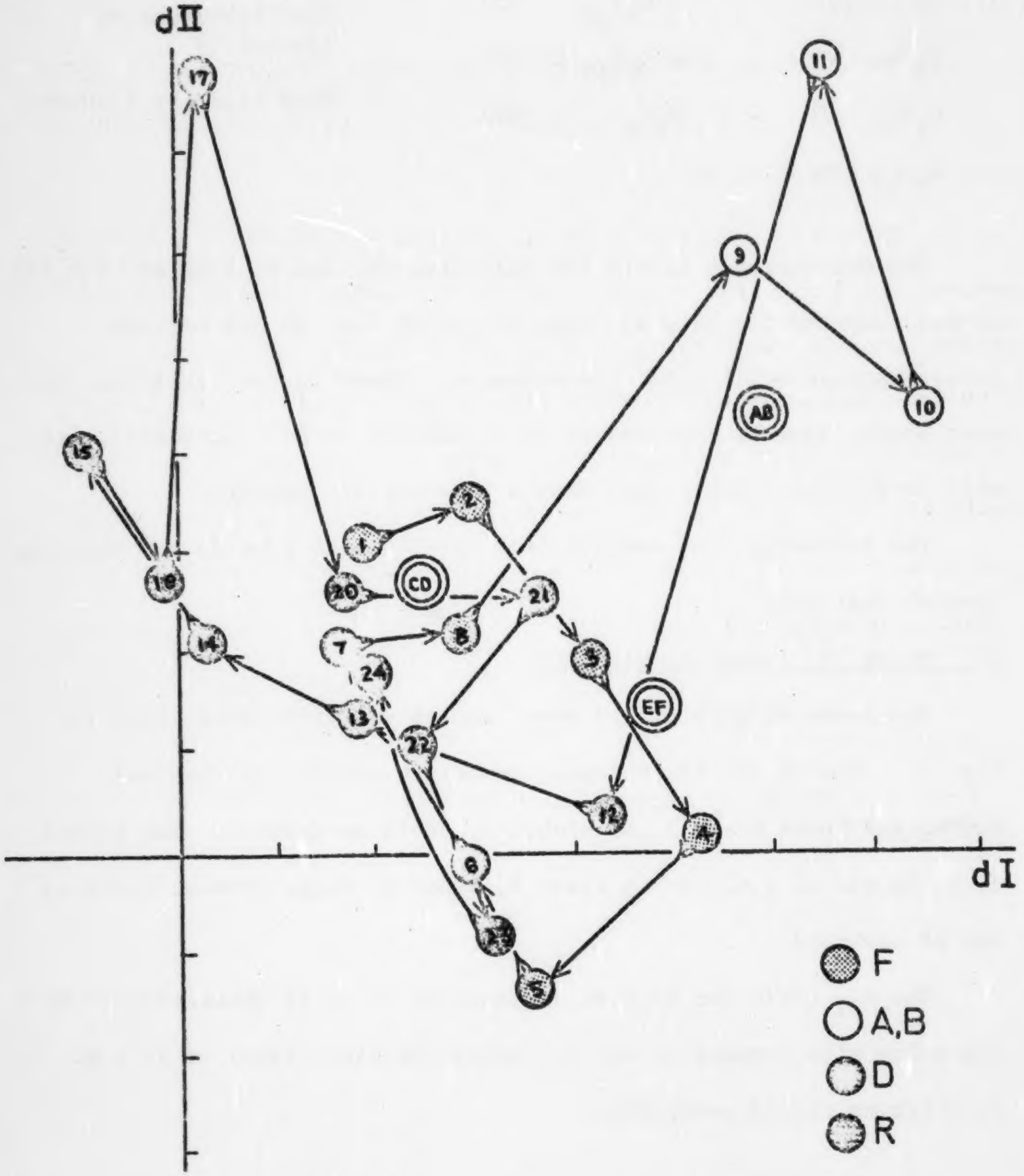
(b) March 13, 1 a.m. to midnight

The score diagram of the hour samples of March 13 is given in Fig. 4. During the first hours, stability class F is observed. During the hours 6 and 7, no stability could be defined. The scores of 9, 10 and 11 a.m. are in class B, A and B, respectively, close to the AB centroid.

The d_{II} score for 12 p.m. (transition to D) is characterized by a low value with respect to the d_{II} score (vertical axis) of 11 a.m. (a shift to the CD centroid).

Fig 4: Score Diagram for March 13, 1974, 1 a.m. - midnight

Score diagram voor 13 maart 1974 1 t/m 24 uur



The following can be concluded from the discriminant analysis:

1. Classifications of the data of multiple components according to stability classes seems significant. One can expect that additional O_3 concentration information will increase the possibilities for classification.

2. The ratio $NO:NO_2$ is generally lower for the stability classes C and D. For the stability classes A and B relatively high concentrations of C_2H_2 (acetylene) are measured.

3. The presentation of data of multiple air polluting components measured simultaneously at one location, for example the Vlaardingen station, improves with the use of so-called score diagrams. The location of an hour sample in this type of diagram can provide an insight in the relation between that sample and other factors, such as atmospheric stability. This location is computed from a data file already classified with respect to that factor. However, the hour sample is not necessarily contained in this data file.

References

1. Hope, K., Methods of Multivariate Analysis, University of London Press, London, 1968.

Figures (Appendix II)

Fig. I	Location of Measuring Stations
Fig. II-VI	Average Concentrations for Each Wind Direction Sector at Vlaardingen for December 1973 and March 1974
Fig. VII	Average Wind Velocity in Knots for Each Wind Direction Sector at Vlaardingen in December 1973 and March 1974
Fig. VIII-IX	Average Concentrations in ug/m^3 for Each Wind Velocity Group for Wind Direction Sector 5 at Vlaardingen (Dec.) Key: knopen = knots
Fig. X	NO/NO_2 Relation as a Function of the Wind Velocity and for Each Wind Direction Sector at Vlaardingen for March 1974 Key: 1. Wind velocity in knots
Fig. XI-XIV	Average Concentrations in ug/m^3 for Each Stability Class for December 1973 and March 1974 at Vlaardingen Key: 1. Stability class 2. March 1974

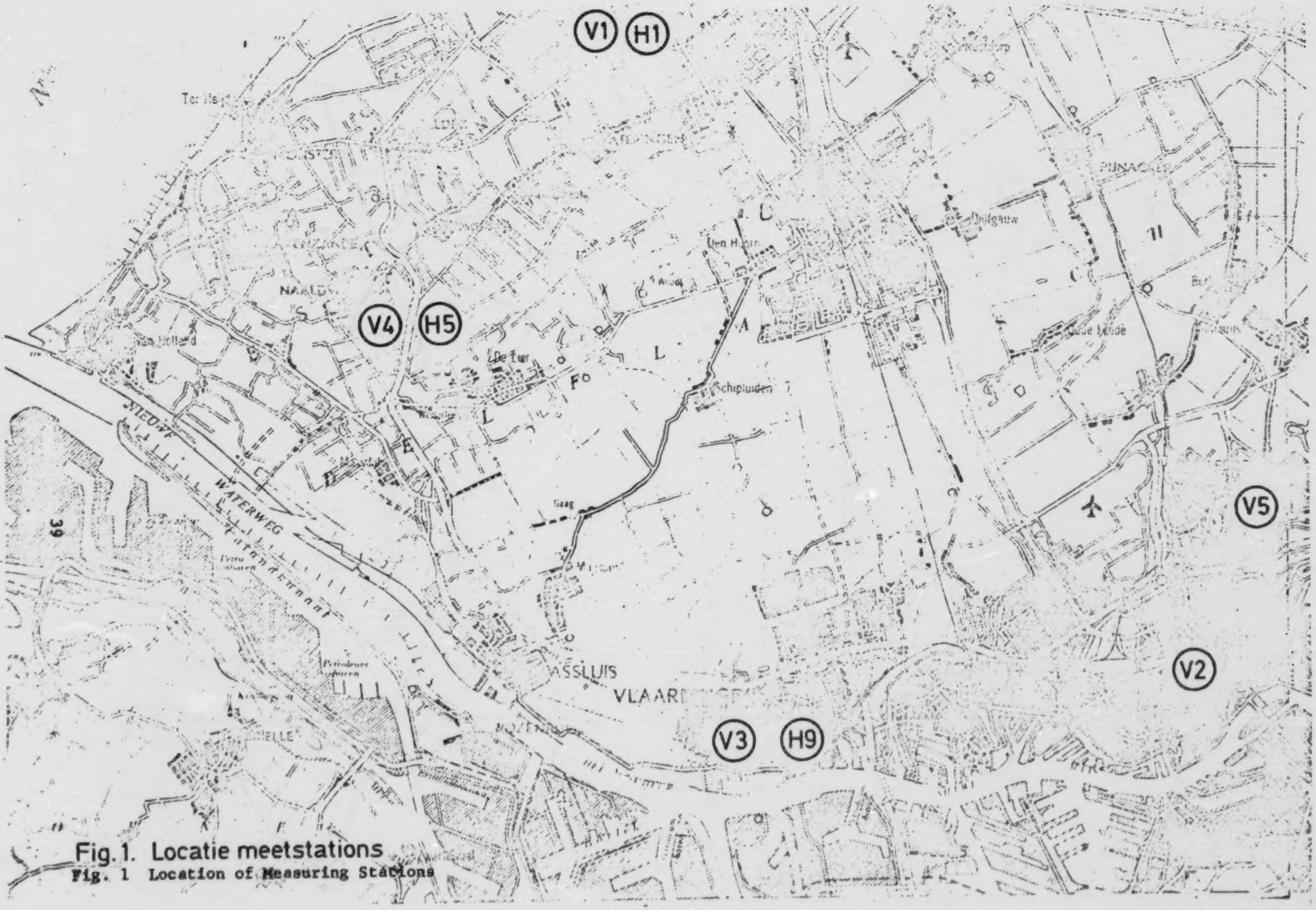

V1
H1
V4
H5
V5
V2
V3
H9
PIJNACKER
Delfgauw
Den Hoorn
Schipluiden
Oude Leede
De Lier
Ter Heijde
NAALDW
NIEUWE
WATERWEG
ASSLUIS
VLAARD
D
E
L
F
L
A
N
D

Fig. 1. Locatie meetstations
Fig. 1 Location of Measuring Stations

Fig. II: Gemiddelde concentraties per windrichtingssector te Vlaardingen over december 1973 en maart 1974.

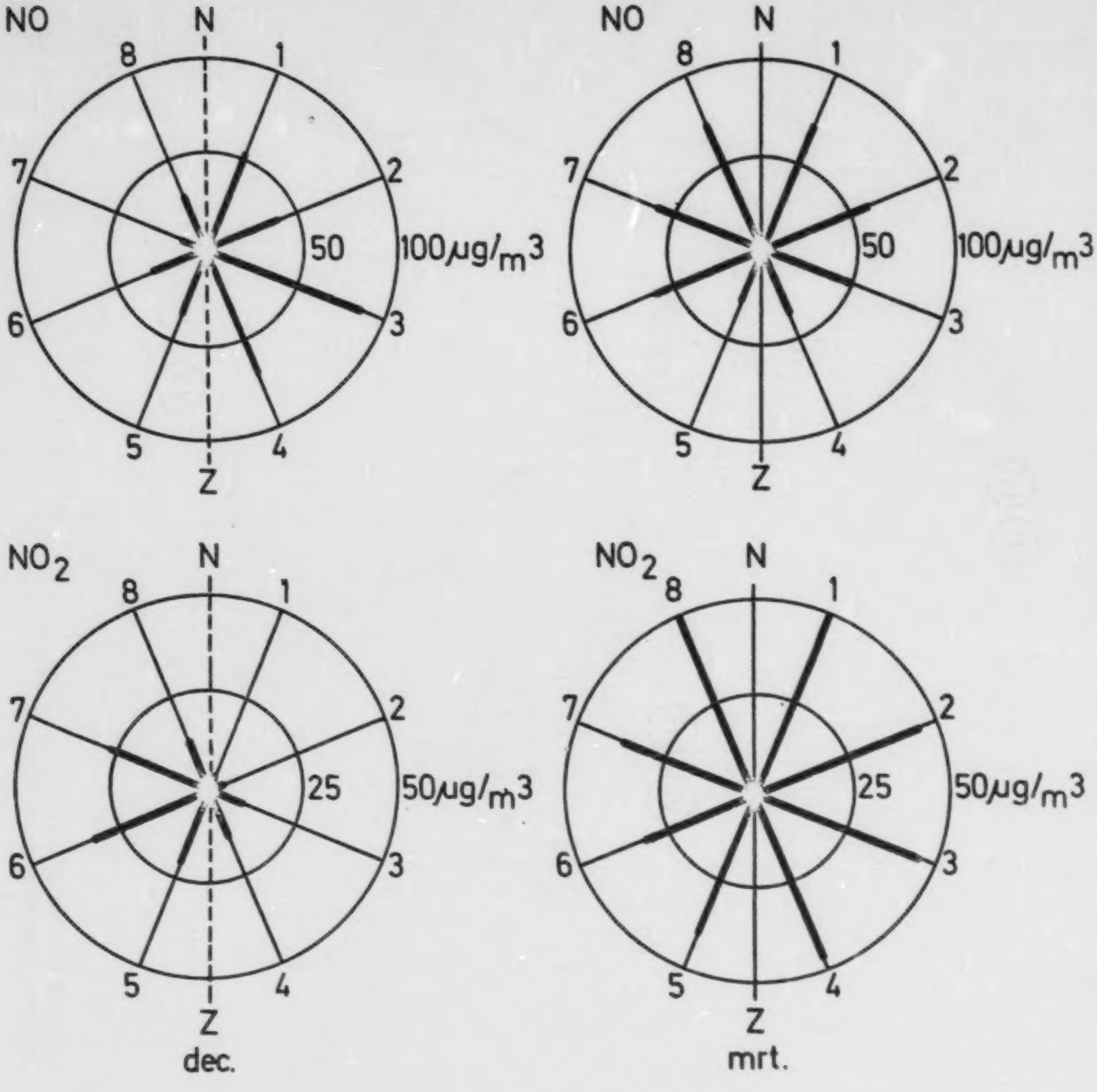
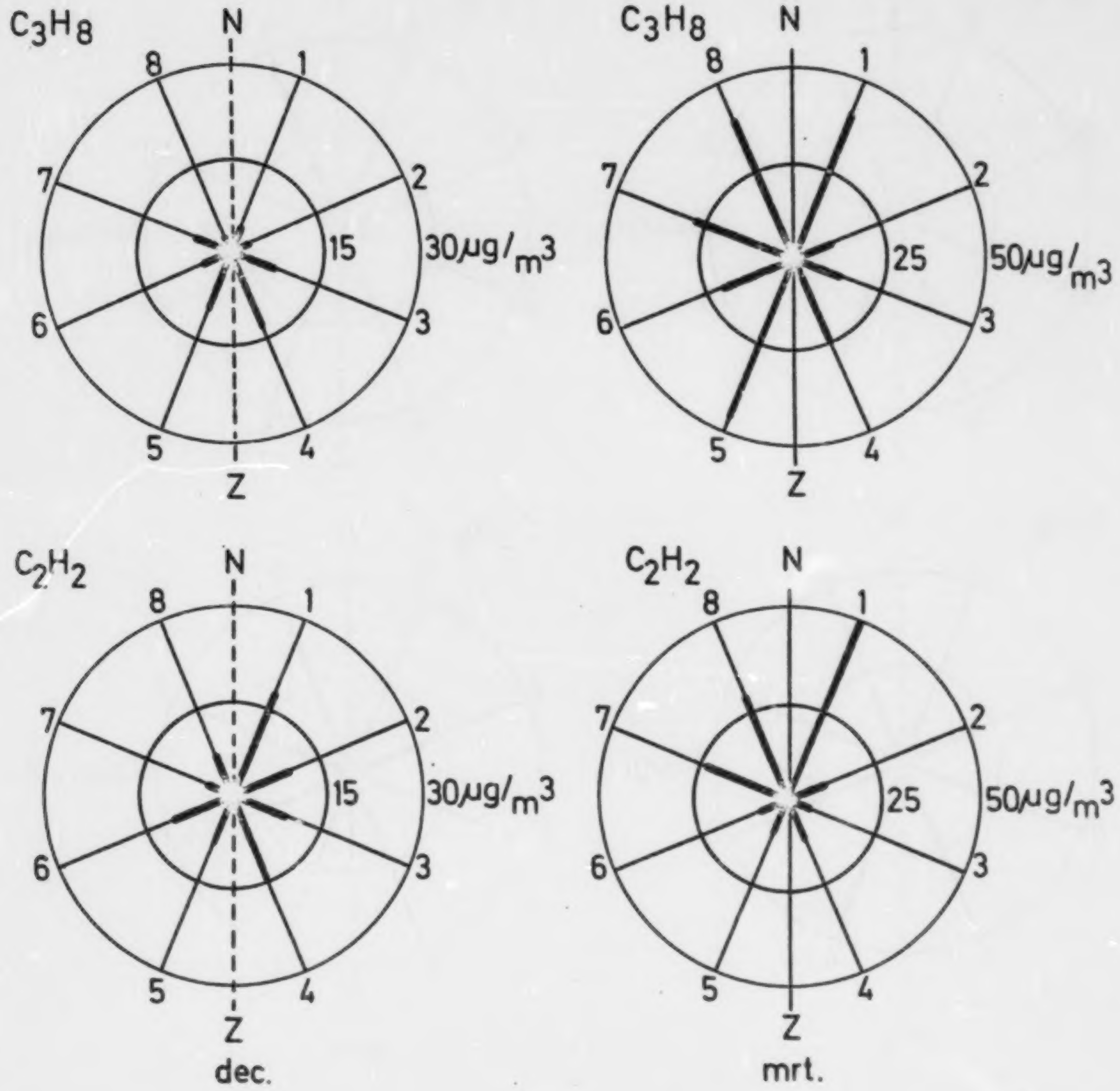


Fig. III: Gemiddelde concentraties per windrichtingssector te Vlaardingen over december 1973 en maart 1974.



dec.

mrt.

Fig. IV: Gemiddelde concentraties per windrichtingssector te Vlaardingen over december 1973 en maart 1974.

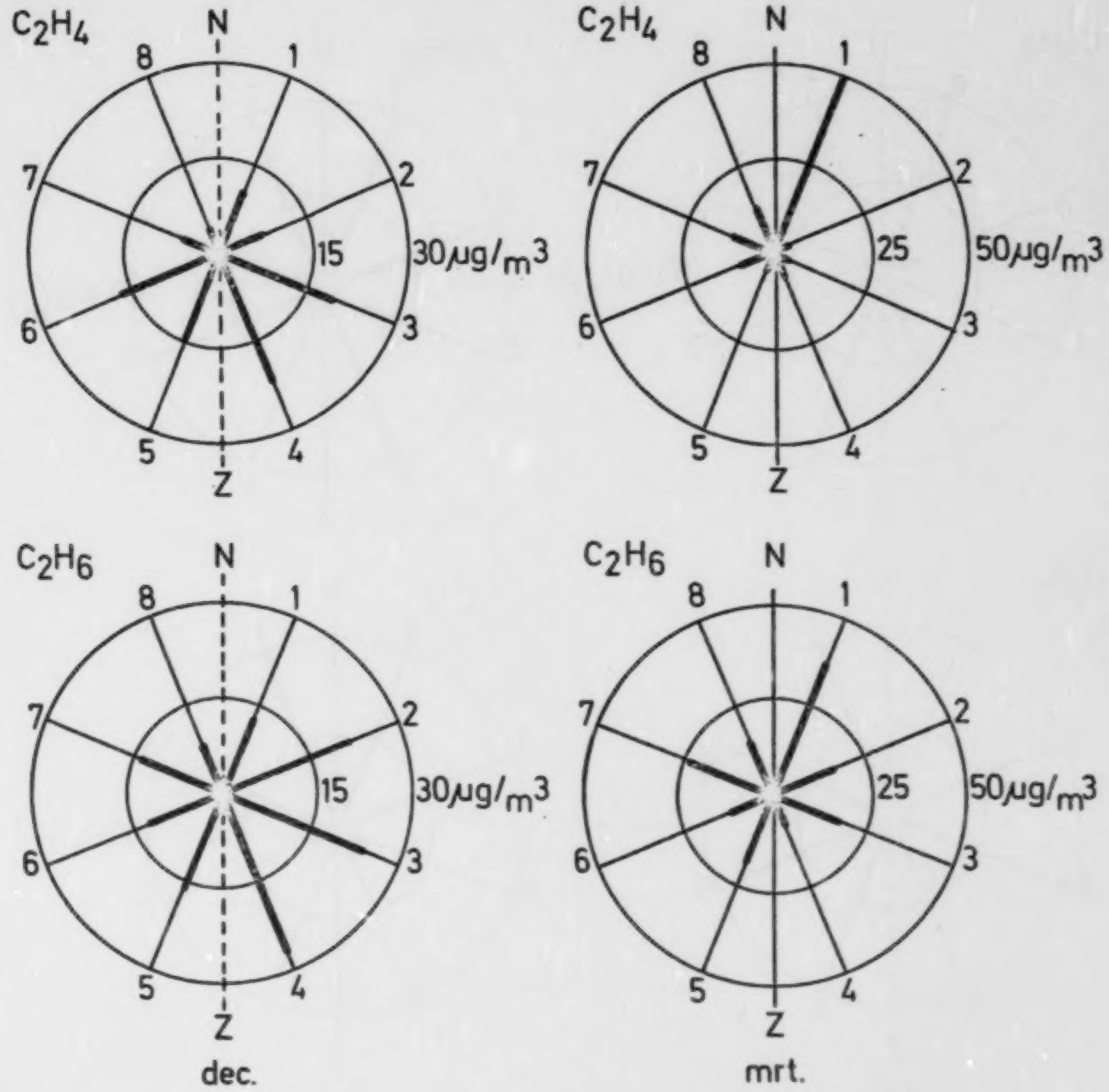


Fig. V: Gemiddelde concentraties per windrichtingssector te Vlaardingen over december 1973 en maart 1974.

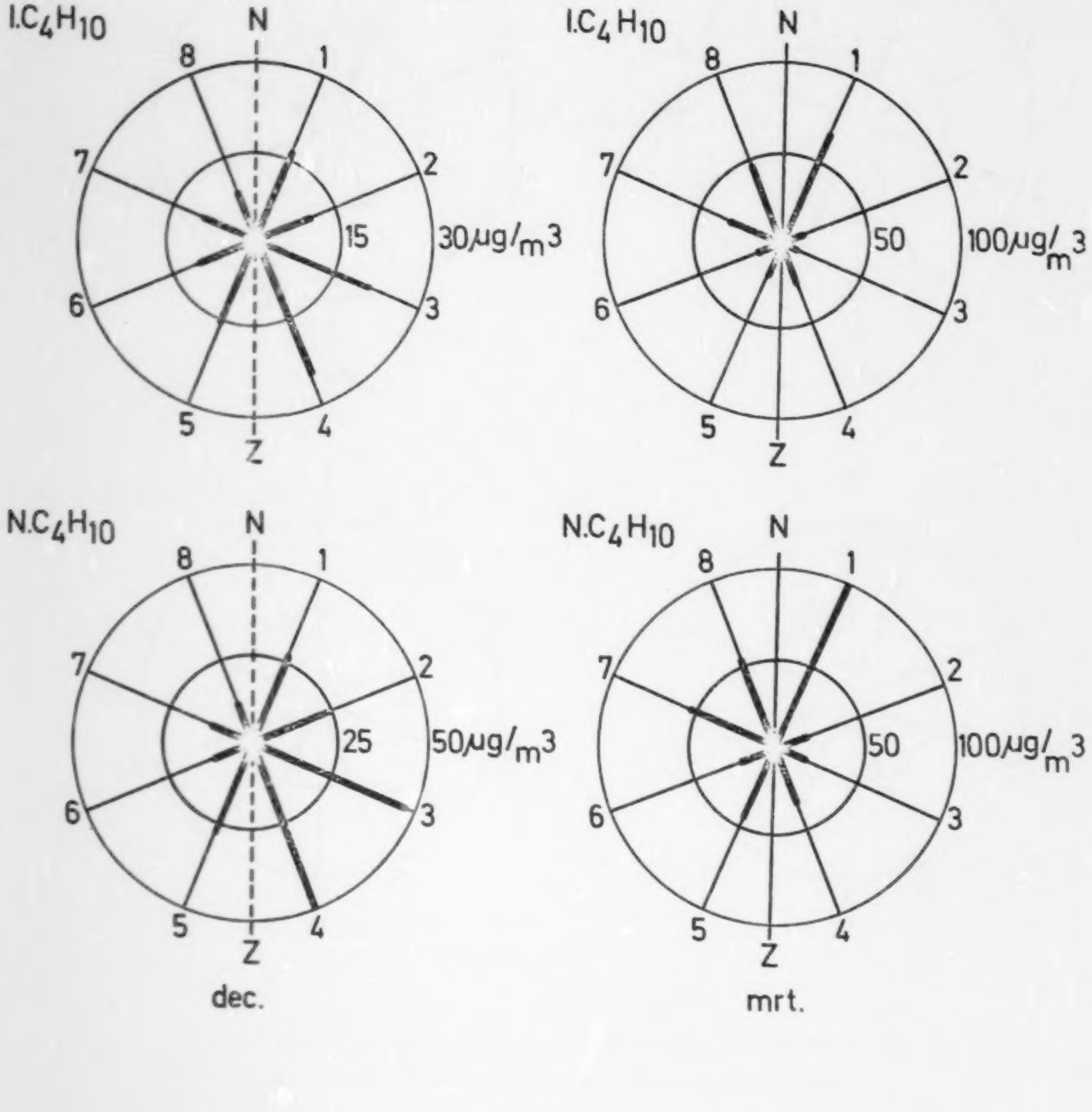
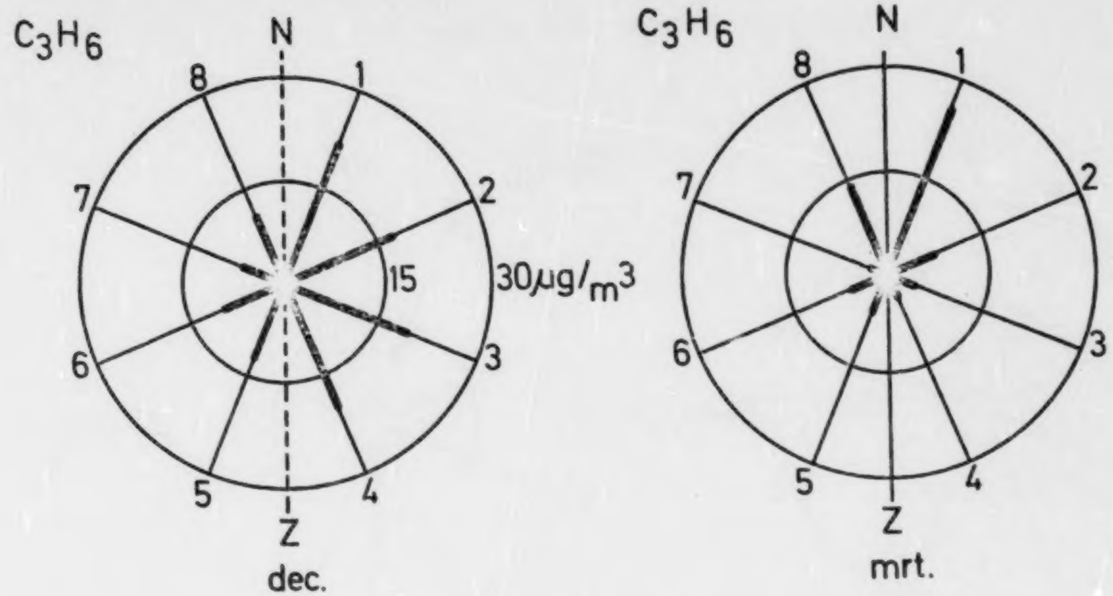


Fig. VI: Gemiddelde concentraties per windrichtingssector te Vlaardingen over december 1973 en maart 1974.



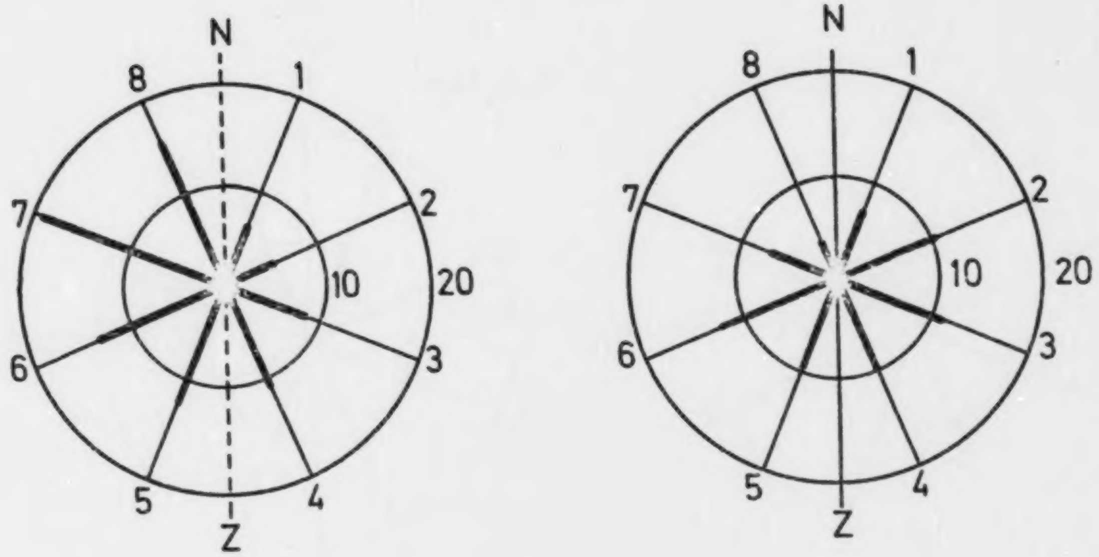


Fig. VII: Gemiddelde windsnelheid in knopen per windrichtingssector te Vlaardingen over december 1973 en maart 1974.

Fig. VIII: Gemiddelde concentraties in µg/m^3 per windsnelheidsgroep voor windrichtingssector 5 te Vlaardingen (december).

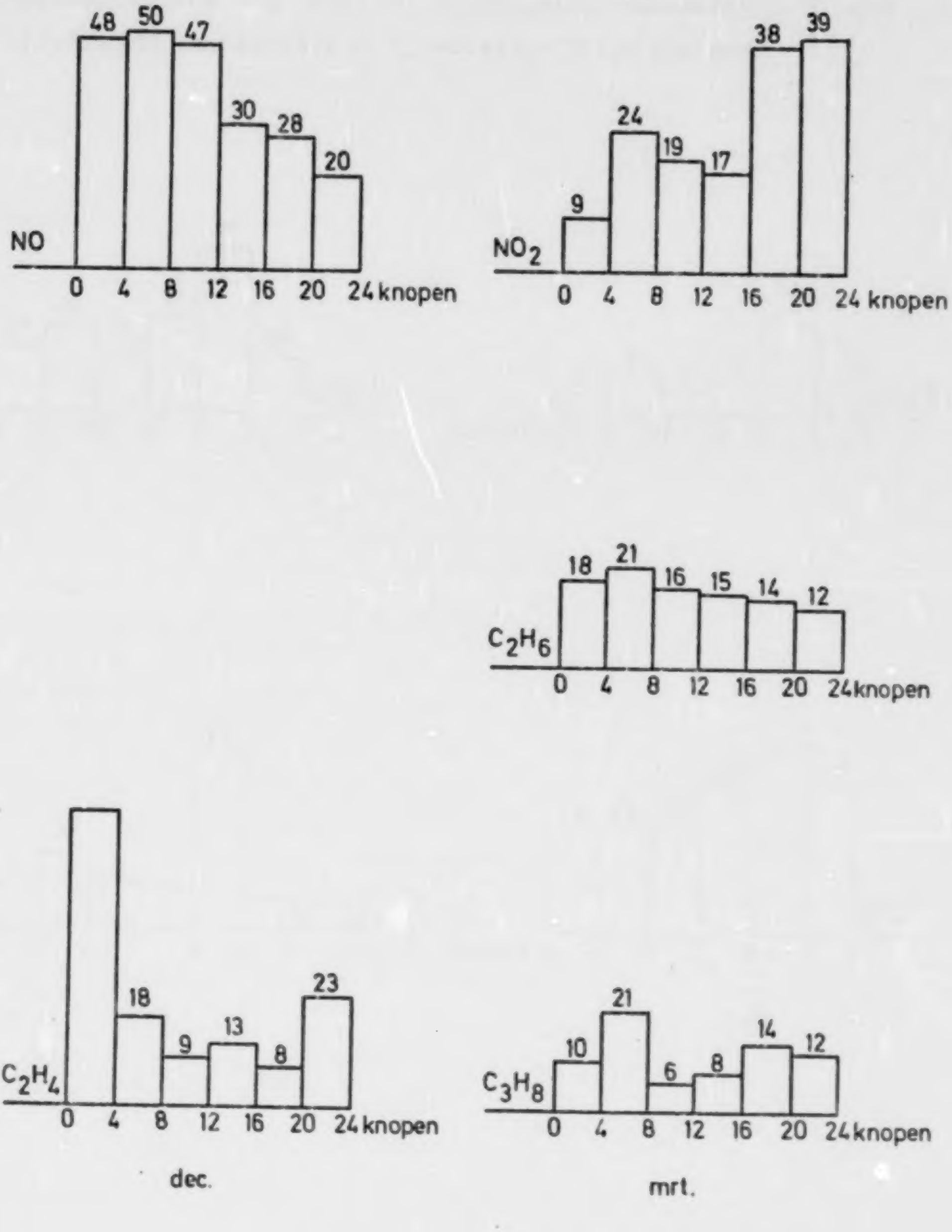
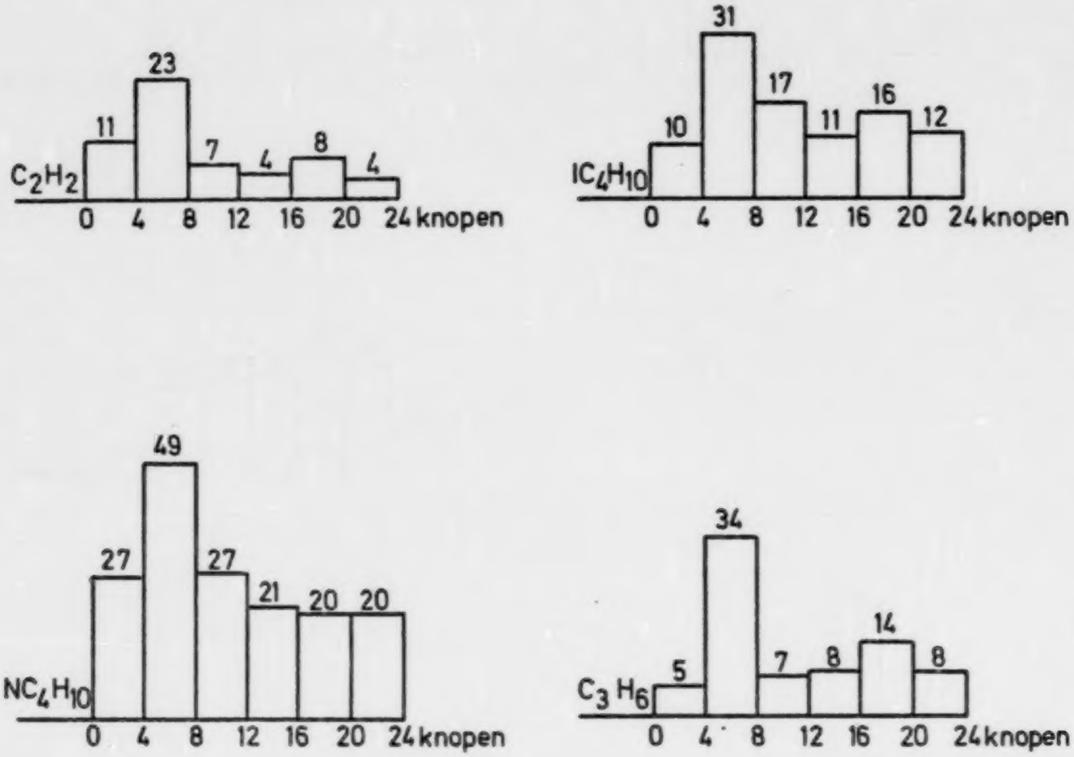


Fig. IX: Gemiddelde concentraties in µg/m^3 per windsnelheidsgroep voor windrichtingssector 5 te Vlaardingen (december).



dec.

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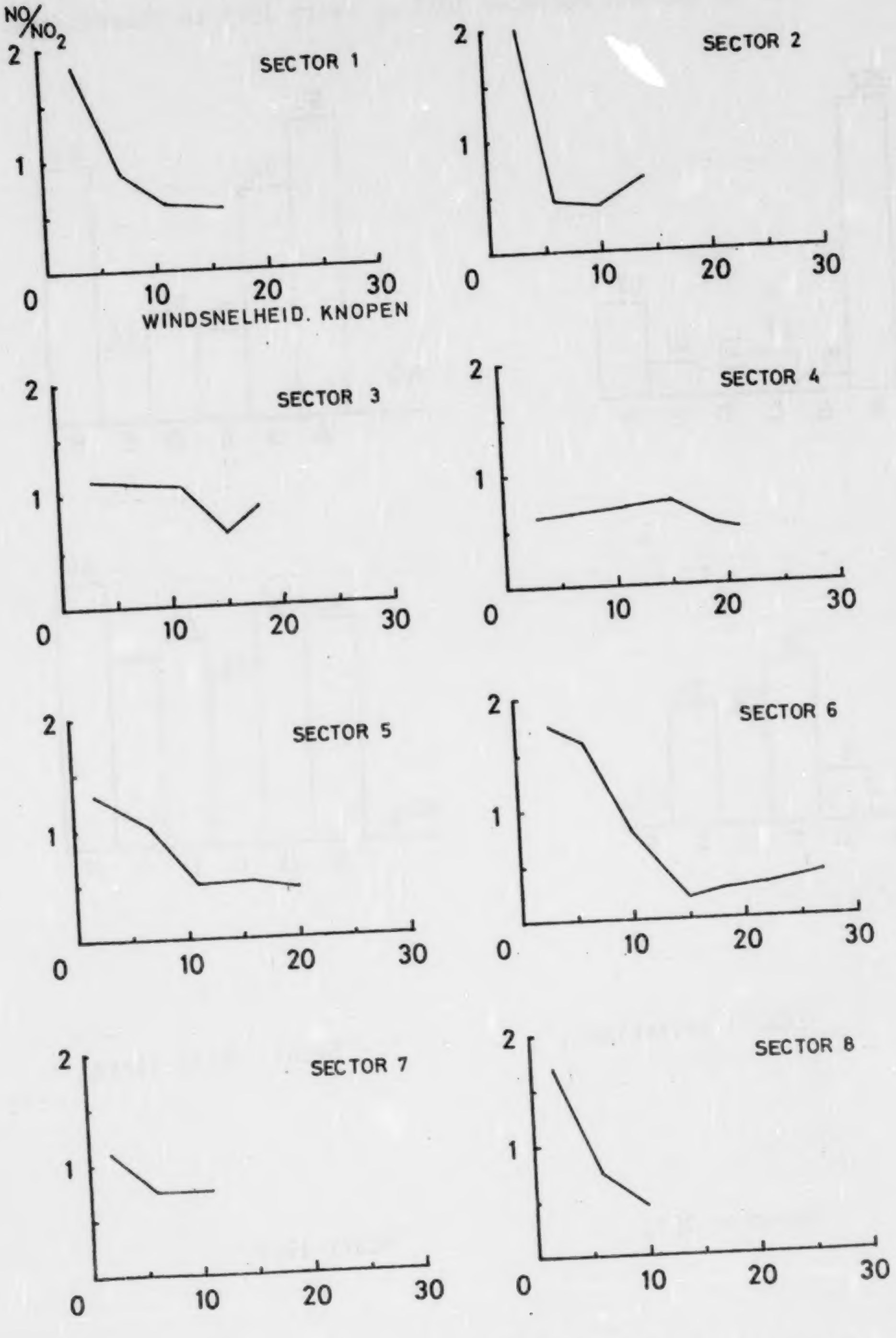
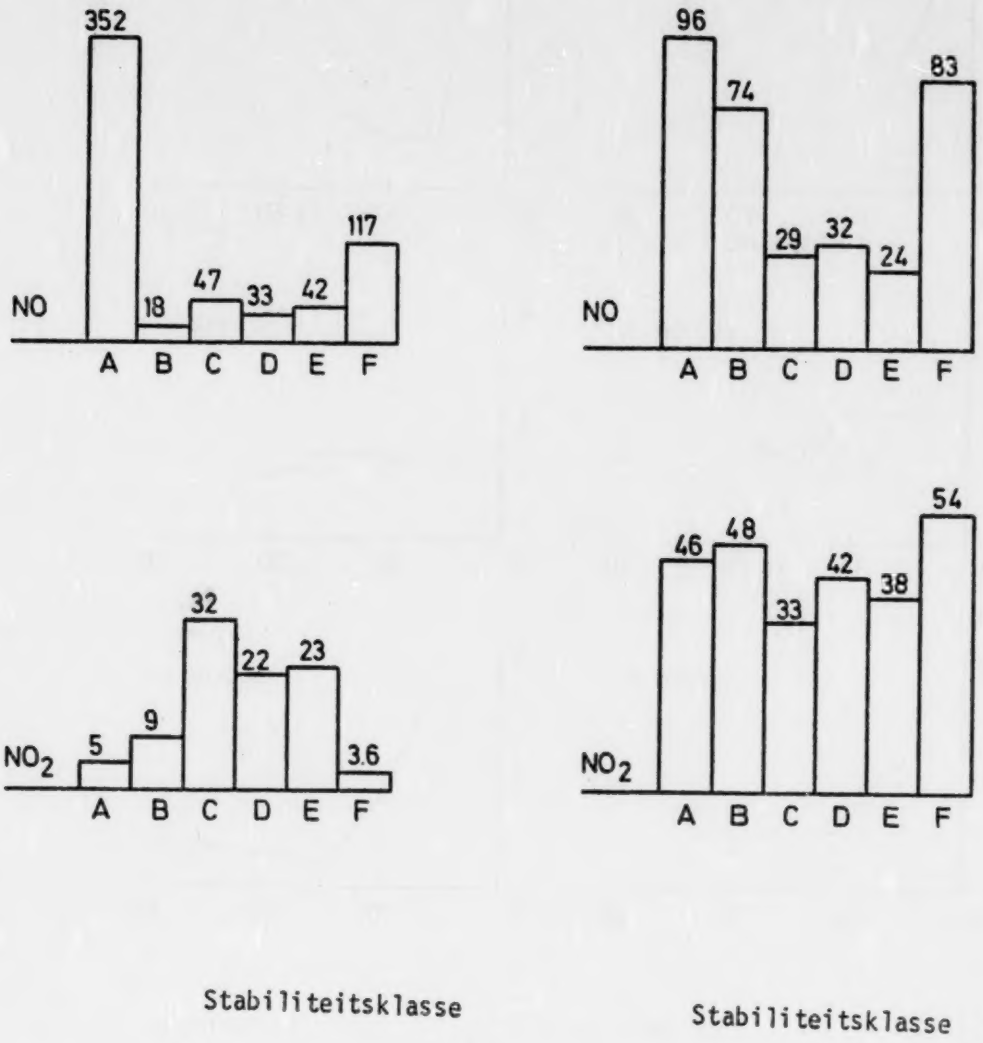


Fig.X : NO/NO$_2$ verhouding als funktie van de windsnelheid en per windrichtingssector te Vlaardingen over de maand maart 1974.

Fig. XI: Gemiddelde concentraties in µg/m^3 per stabiliteitsklasse voor de maanden december 1973 en maart 1974 te Vlaardingen.



December 1973

Maart 1974

Fig. XII : Gemiddelde concentraties in $\mu g/m^3$ per stabiliteitsklasse voor de maanden december 1973 en maart 1974 te Vlaardingen.

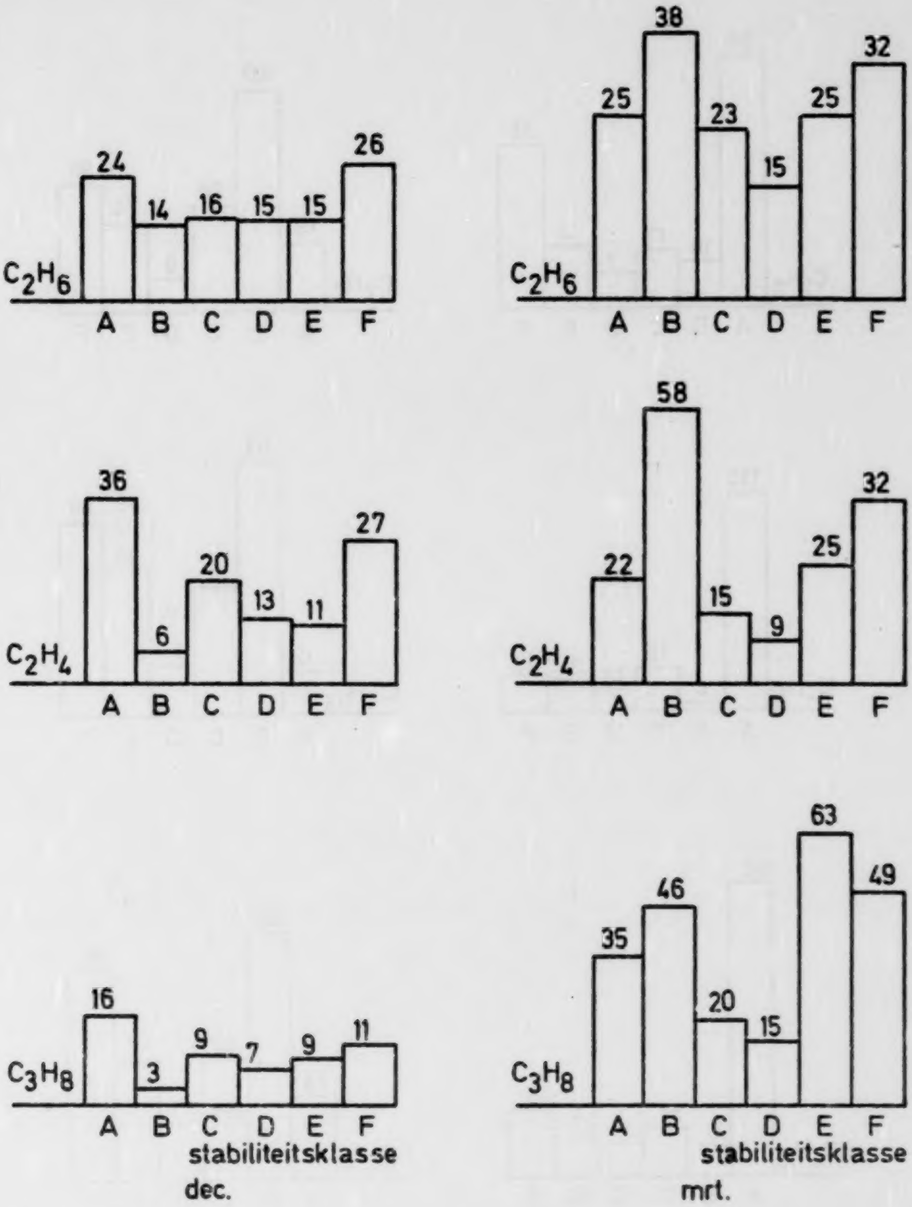


Fig. XIII: Gemiddelde concentraties in µg/m^3 per stabiliteitsklasse voor de maanden december 1973 en maart 1974 te Vlaardingen.

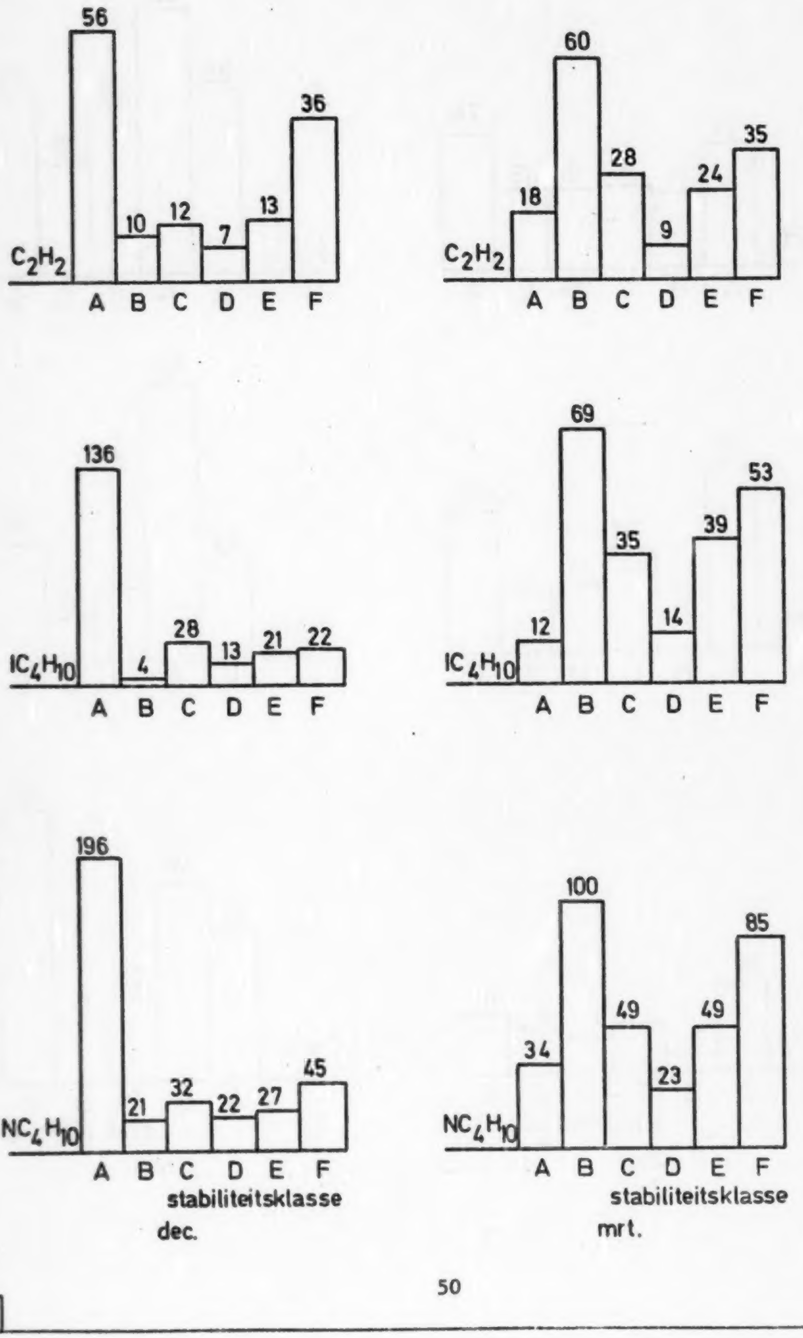
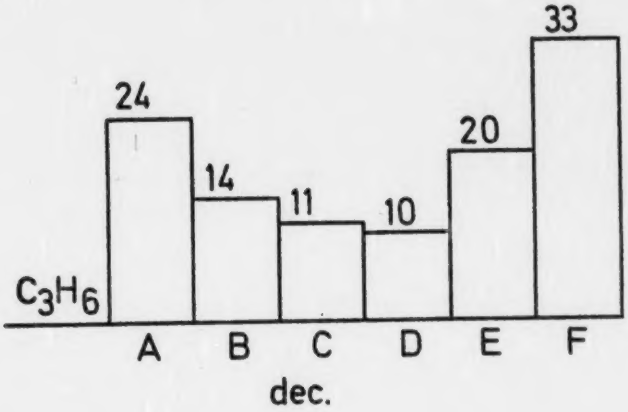
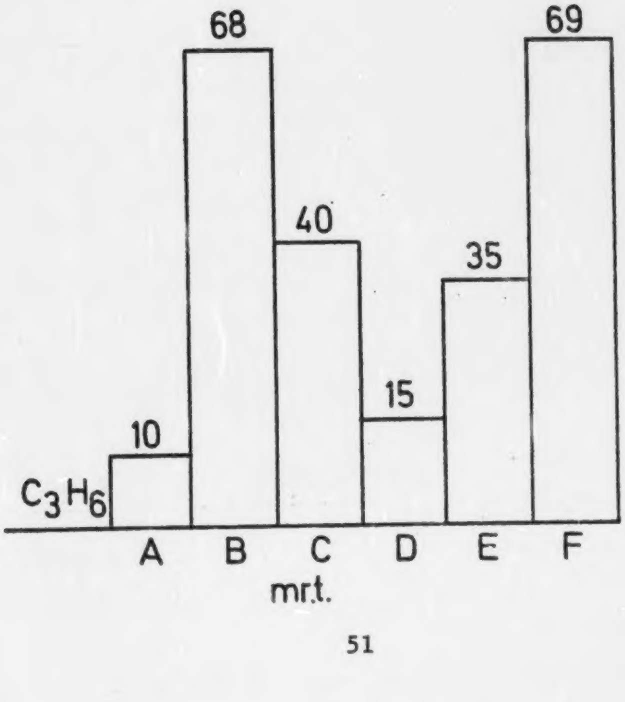


Fig. XIV: Gemiddelde concentraties in µg/m^3 per stabiliteitsklasse voor de maanden december 1973 en maart 1974 te Vlaardingen.





APPENDIX III

Overview of the Investigations into the Influence of Air Pollution on Plants for the "Proefprojekt"

Research Institute for Plant Diseases (IPO)
Wageningen

Introduction

From the start of the air pollution research into the effects on plants (1954), the IPO has paid attention to the Nieuwe Waterweg region. This investigation was initially concentrated around Pernis and Hoogvliet and the region between Vlaardingen and the Westland. In the first region every year a certain amount of leaf damage in the apple and pear orchards had occurred. In the second region damage by air pollution containing fluor was observed already before 1954. Gradually the investigation was extended to the entire Westland; in later years the region south of the Nieuwe Waterweg was added, too.

Research 1973

A. Field Investigations

The indicator plants for the air polluting components HF, SO_2, O_3, NO_2, PAN (peroxyacetylnitrate) and ethylene were not cultured anymore in the field but in uniform soil in containers with constant moisture level maintained by a water siphon system.

The indicator plants showed the following: Spinach (sensitive to SO_2 and O_3) showed damage in May in the region south of the Nieuwe Waterweg, especially between the 14th and 27th of May. This might

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correlate with O_3-, SO_2- and $O_3 + SO_2$- concentrations measured in this period for this region. Petunia (sensitive to PAN) shows a symptom characteristic for PAN in the period July 2-22 and between September 10-16. Small stinging nettle and street grass (sensitive to PAN) were damaged in the period September 10-23. The nettle was also very lightly damaged between July 2 and July 18, and also between August 20-26. Right before September 10th, the Central Laboratory TNO at Delft measured PAN at Delft. They observed a definite PAN damage to the small stinging nettle, street grass and Petunia at Delft (test fields at station IG-TNO). A higher than average damage was observed for tobacco BEL W3 in the periods June 18-24, August 27 - September 2, and September 10-16. This observation was based on little damage in the preceding period followed by heavy damage of the same plants in the following period. HF did not offer any new insights compared with other years. The gladiolus and freesia cultured at the test field showed that the HF originated from the Vlaardingen region. In this region the plants in containers showed the heaviest leaf damage. The damage gradually increased. No peak values were seen for specific periods, which is logical in view of the cumulative action of this gas.

B. Research in Small Greenhouses

At six locations in the Nieuwe Waterweg region two greenhouses are placed; one has a carbon filter to filter away the air pollution; the other one lets in the unchanged outside air. The station at Naaldwijk investigated three types of carbon. The first one is the original carbon (type Norit RLI); the second one (type Norit RBAAL) is for removing SO_2 and HF; the third one is not a commercial product but is delivered upon special request and is meant for filtering the ethylene from the air. Measurement of the ozone and nitrogen dioxide contents before and after the filter type RLI showed that all ozone and almost all NO_2 is filtered away. The ethylene filter satisfies well for ethylene; however, this shows only after one month of use. The SO_2 filter satisfied for SO_2 from the start.

The research with filtered greenhouses at Naaldwijk shows that with use of the carbon type RBAAL after some time the plants have grown better than with the use of carbon type RLI. An explanation might be that the first type of filter not only removes the damaging action of ozone, but also (the damaging effects of) the synergistic action of $O_3 + SO_2$ and of $SO_2 + NO_2$. It was also found that in general the tomato and tobacco plants did better in the filtered than in the unfiltered greenhouses. However, there are slight differences in climate that might partially explain the growth differences between the greenhouses with and without filter. At all six locations tobacco was damaged by ozone in the unfiltered greenhouses, while tobacco was completely undamaged in the filtered greenhouses.

In the period March 26 - April 8 the tulips at Naaldwijk, Delft, Maasdijk and Vlaardingen in the unfiltered greenhouses were severely damaged; no damage was observed in the adjacent filtered greenhouses. At the other two locations, Den Haag and Hellevoetsluis, no damage of the plants was observed. The tulip symptoms indicated SO_2 damage, although at the six locations the carbon filter type Norit RLI, the filter without preparation of the carbon for SO_2 removal, was used.

C. Research at Wageningen

A PAN gas application research was started after advice was obtained from the Central Laboratory TNO (Delft) because of the reactivity of the gas. The cylinders with PAN (± 300 dpm in N_2) were also delivered by this laboratory. The research was directed toward the testing of the severe lettuce damage of October, 1965, in the greenhouses of the Westland. This damage was accompanied by the specific damage of the PAN-sensitive plants, the small stinging nettle and street grass. In order to test this phenomenon in the Westland greenhouses, lettuce, small stinging nettle and street grass were included in the PAN gas application. It must be considered that it was not yet possible in 1973 to measure with the purchased gas chromotograph the PAN concentration in the greenhouse.

The gas application test showed that the lettuce used in the Westland was not particularly sensitive to PAN compared with the indicator plants used, small stinging nettle and street grass. The order of PAN sensitivity was: small stinging nettle - very sensitive; street grass - slightly less sensitive; lettuce - much less sensitive. This seems to contradict the information from American publications. The type of lettuce that is known in the USA for its PAN sensitivity is romaine lettuce and is quite different from the lettuce grown in The Netherlands. The gas application tests in 1973 at Wageningen showed meanwhile that romaine lettuce is more sensitive to PAN than Dutch types of lettuce. Four types of romaine lettuce were tested; one type showed extreme sensitivity to PAN.

Because romaine lettuce can be grown in The Netherlands in the fields during the summer, this lettuce will be used in the fields as an indicator plant for PAN. Although the test was negative with respect to the observed damage of the Westland lettuce at the beginning of October, it is still very likely that the damage of this year was caused by PAN. This was shown by an intensive inspection of this region in October, 1970. In the autumn of 1970 specific PAN damage of the small stinging nettle, street grass and young lettuce plants were observed again. We can classify the surrounding region with respect to the industrial Botlek region as follows.

At the greatest distance (West-Gaag, Oost-Gaag) only damage of the small stinging nettle which grew as a weed at several places in the greenhouses was observed. No lettuce was damaged.

The next zone: the nettle was quite damaged in the greenhouses, street grass slightly damaged; no lettuce damage.

Zone 3: nettle strongly damaged, street grass less, cultured lettuce slightly damaged.

The zone closest to the industrial region where greenhouses are in use: nettle and street grass strongly damaged, various other wild plants less damaged, lettuce strongly damaged.

The PAN gas application tests show the same pattern; in both cases the indicator plants showed the same symptoms. It must be concluded that for a large region severe PAN damage has occurred in the Nieuwe Waterweg region in October, 1965; in October, 1970, a smaller region was damaged. This indicates that a regular PAN concentration measurement in the Nieuwe Waterweg region would be desirable.

A gas application test with ozone is also carried out. Tobacco (cultivar Bel W3) was used as an ozone indicator plant in the field and the greenhouses. Because of the great ozone sensitivity of Bel W3, it was not possible yet to obtain reproducible data.

APPENDIX IIIA

Gas Applications of Peroxyacetylnitrate (PAN) to Plants in 1973

Research Institute for Plant Diseases, Wageningen

In the winter of 1972-73, ten types of lettuce grown in the Westland region were tested for their PAN sensitivity by gas application in small greenhouses especially built for this test. For these tests, the minimal concentration for which leaf damage still could be observed was 300 ppb PAN during a 3 hour exposure. (Note: the concentration is computed, not measured.) According to the literature, damage can already occur with a 30 ppb PAN concentration. For this reason new tests are indicated to investigate the sensitivity of lettuce to PAN:

A. In the summer the American romaine lettuce, which according to the literature shows damage already for a 30-50 ppb PAN concentration, was tested.

B. The tests during the winter of 1972-73 with Westland lettuce, and during the summer of 1973 with romaine lettuce, were followed up in the autumn of 1973 with a test of three varieties of lettuce.

C. In addition, the small stinging nettle (Urtica urens) was tested in the autumn of 1973.

A. Test Set-Up and Results of PAN Gas Applications to Romaine Lettuce

The following varieties of romaine lettuce were tested for their PAN sensitivity: '68206', '68853', '69010' and '69854'. The seed was obtained from the "Instituut voor de Veredeling van Tuinbouwgewassen" in Wageningen.

Set Up

The plants are germinated with a night temperature of 13°C and a day temperature of 17°C in a climate cell. After 11 days (1 heart leaf) they were planted out and put in a cultivation room with a day temperature of 25°C, an RV (relative humidity) of ± 50%, and a night temperature of ± 15°C with an RV of 70-80%. The gas was applied in June. The plants, in a stage of 5-6 leaves, were exposed to the PAN gas for a few hours. During the gas applications the temperature and the RV could not be controlled because the old system was still in use. The information was registered by a thermohygrograph. The temperature during the various gas applications was linked to the weather conditions and could rise as high as 35°C, with an RV varying between 50-70%. The RV remained quite stable during each gas application. The PAN concentration measurements with the gas chromotograph was troublesome. The given concentration is computed from the air volume (liters/minute) to which a certain amount of PAN/N_2 mixture of known concentration was added. During the gas application the light source was a set of fluorescent light tubes, ± 20,000 lux. After PAN exposure the plants were returned to the cultivation room.

Observations

A concentration of 100 ppb PAN for three hours caused in most cases a visible damage. The checking was done six days after the gas application. Visually the percentage of damaged leaf was assessed. The extent of the damage was different for each plant variety. The back of the leaf was damaged more severely than the upperside. Most of the time a part of the leaf showed a silver gloss, sometimes the front side of the leaf, too. This pattern was the same for all varieties.

Three hours after the gas application the damage was visible already: a watery, shiny damage at the back side of the leaf. This disappears later and causes light green stains or, for serious damage, bronze colored stains. The so-called band formation was clearly visible. The oldest leaves were damaged at the base of the leaf; the younger leaves, in the middle; and the youngest,at the top of the leaf. Mature leaves and young leaves remained undamaged. The following damage pattern could be observed after one day:

In the case of light damage, light brown to bronze colored spots along the edges of the leaf were observed. In the case of more serious damage the tissue is dead, sometimes green and droopy, mostly light brown to bronze colored. The main rib and the side ribs are mostly intact. Gloss is observed at separate spots or at the edge of a damage band (band formation).

The Results

The results are given in Tables I and II.

The leaf is judged at the backside.

The hours of sunshine and the global radiation are data obtained from the weather station of the laboratory of Physics and Meteorology at Wageningen.

Explanation of the symbols and abbreviations:

68853, 69854, 68206, 69010: the varieties of romaine lettuce

+ : the number indicates the total percentage of damaged leaf surface of the damaged leaves

- : indicates the percentage of glossy leaf surface

the amount of light: the amount of light measured at the day of the gas application

gs: no damage

B. Set Up and Results of the Autumn Gas Application Tests of Three Lettuce Varieties

The varieties 'Amanda +', 'Miranda' and 'Deci minor' are selected from the ten varieties used previously in gas tests. The selection was guided by the following factors: cultivation of these varieties at the Westland region, the time of the year, and the plants' reactions at earlier gas tests. Purpose of the test was to detect whether PAN sensitivity changed with time because of differences in the length of the days in the months of September, October and November. The day length defines, among others, the velocity of growth and therefore probably sensitivity too. The amount of light before and after the gas application will be greater in September than in November, which might cause a more severe leaf damage. In order to exclude sensitivity differences caused by the development stages of the plant as much as possible, each week a new series was started. The use of young plants provided homogeneous, fast growing material. Other advantages of the use of these young plants were:

1. In addition to the visual inspection of the damage there was an opportunity now to compute the damage by measuring the ratio of dry

and fresh weight. It was assumed that the fresh weight of the treated plants would be lower than that of the untreated plants because, in most cases, leaf damage causes the tissue to die; this will dry and cause a lower percentage of fresh weight. Thus, the percentage dry weight/fresh weight would be higher for treated plants. This method can still be used for young plants because almost all leaves are in the sensitive growth stage, as opposed to the older plants with mostly insensitive leaves.

2. The small plant size allows treatment of several plants in one variety at the same time. Limiting factors are the size of the used gas application greenhouses and the boxes procured for the culture of the plants. The size of these boxes (13 x 20 x 30 cm) made it possible to treat at least three plants of one variety at the same time.

The Set Up

The plant material was obtained in the form of plants in pressed pots ("Jiffy" pots) from a nursery. Six plants were potted in a plastic container, two times three of each variety. The containers were filled with ± 7 liters of sandy soil from the test station at Naaldwijk; the plants were given extra nutrients as advised. In these containers porcelain candles were installed which regularly could moisten the soil. The use of these containers made it possible to compare the reactions of the plants because of the uniform water supply, soil and location. The most important point is the uniform water supply during the gas application. The plants were cultivated in a room with a night temperature of ± 10°C and a day temperature of ± 15°C, which increased with sun up to ± 20°C. The relative humidity RV was 70-80%. The plants were treated for three hours with ± 175 ppb PAN or for four hours with ± 75 ppb. The three-hour gas application took place from 9-12 a.m. or from 1-4 p.m. The four hours were from 11 a.m. - 3 p.m. During these gas applications the temperature and RV were dependent again on the weather. The data were registered by a thermohygrograph. The light intensity during the treatments was 15-20,000 lux. The wetness of the soil could be maintained at a constant level with the aid of the capillary action of the candles. The tiny supply tubes of the candles floated in a tray with a water level kept constant with the aid of a barrel outside of the greenhouse. The barrel was kept filled with an upside-down bottle. The PAN concentration could not be measured but is computed (see also A above). After the treatment the plants were returned to the culture room; the amount of light and the light intensity here were dependent on the weather condition.

Observations

The observed damage and the symptoms of the damage were almost identical to the earlier described symptoms of romaine lettuce. Sometimes no necrose or gloss was visible on the sensitive places of the leaf; a yellow coloring was observed instead.

Results

The results are given in Tables III and IV.

Table III: The total damage of a plant, that is, the sum of the percentages of leaf damage per plant is given. The averages per treatment, per three plants are given.

Table IV: The percentage dry/fresh weight per series, per duration of application. For each treatment three plants are used.

Explanation of symbols and abbreviations for Table III:

Damage: the sum of the percentages of leaf damage for the three varieties, per plant
A: the variety Amanda+
M: the variety Miranda
Dm: the variety D minor
Begas. tijd: the duration of the treatment
Uren zon: hours of sunshine after the end of the treatment (because this factor can influence the effect of the damage). The data were obtained from the laboratory for Physics and Meteorology at Wageningen.

It is hard to produce PAN in big quantities; PAN is also semi-stable, and therefore only small amounts could be obtained at a given time. Because of difficulties with the synthesis of PAN at the Central Laboratory TNO at Delf, the tests had to be stopped prematurely. More repetitions of the gas application would probably have provided more information about the PAN sensitivity of lettuce plants.

C. Gas Applications of the Small Stinging Nettle (Urtica urens) with Peroxyacetylnitrate (PAN)

The plants were grown in the same containers and under the same weather conditions as the lettuce plants in the autumn experiment described earlier. Eleven days after sowing, the plants were planted out and twelve days later repotted. They were treated simultaneously with the lettuce plants and had a thin, protracted and irregular shape. Treatments were three hours of 175 ppb or four hours of 75 ppb. The three hours were from 9 a.m. - noon or 1 - 4 p.m., and the four-hour treatment was from 11 a.m. - 3 p.m.

Observations

A great variety in the extent of the damage for the different plants was observed. However, the pattern of the damage was uniform. A day after the gas application the leaf was spotted dark green, later light green up to brown. The final damage is a white necrose. The front of the leaf showed more damage than the backside of the leaf. The ribs and stinging hairs were intact in most cases. A so-called band formation was visible after all treatments, that is, a band of damaged tissue extending over the width of the leaf. For the oldest leaves this was at the base of the leaf, for the younger ones at the middle and for the youngest leaves at the top of the leaf. For serious damage in the middle or the base of the leaf, the top still stayed green because the main rib was saved. Light damage, a band of 2-4 mm, starts at the edge of the leaf in the oldest part of the leaf and from there extends by way of the teeth to the younger part of the leaf; the parts join at the top or the middle of the leaf. This arc shaped leaf damage only damages the leaf pulp along the ribs and around the stinging hairs.

Results

A great variety in the reactions of the nettle to PAN gas applications was observed. A few times no damage was observed after a three hour, 175 ppb treatment. On the other hand, serious damage was observed sometimes after a four hour 75 ppb treatment. In general the small stinging nettle will become seriously damaged after three hours of 175 ppb PAN gas application. A 75 ppb (four hours) treatment caused damage too, while the lettuce, treated simultaneously, showed no or very little PAN damage symptoms.

Remarks:

1. The given concentrations are computed, not measured.
2. To obtain a correct picture of the PAN sensitivity of the small stinging nettle, a relatively great number of gas applications have to be performed. This was made impossible by the stagnation in the delivery of PAN cylinders. The impossibility of concentration measurements is another factor that supports an extension of the tests.
3. Plants of normal habitus should be used for the gas applications.

The Ozone Gas Applications to Bel W3 Tobacco

In the spring a preliminary test was set up to test whether the tobacco Bel W3's sensitivity was dependent on the age of the plant or the season.

The plants were grown in pots in a culture room with a night temperature of ± 18° C, day temperature of 20° C, increasing up to 25° C, and a RV of 65-75%. The plants obtained additional light from HPL lights from 8 a.m. - 8 p.m. The light intensity was 10,000 - 20,000 lux (dependent on the distance from the pot to the light).

For acclimatization the plants were placed in the gas application greenhouses with aeration the night before the treatment. The air velocity was ± 4.40 m^3/min; that means for a soil surface of 2 x 2 meters, an air displacement of 1.5-2 cm per second. The temperature was ± 19°C and the RV 70-80%. In the morning the light was switched on in the gas application greenhouse, and the plants were watered. At 10 a.m. the gas was applied until 4 p.m. The temperature during the treatment was 20-24°C and the RV 70-80%. The desired ozone concentration was ± 100 $\mu g/m^3$.

An ozone monitor was not available for all treatments. To make an evaluation possible, both the percentage of leaf damage and the velocity of growth were registered weekly.

Results

The series and also the plants showed great relative differences in their reactions to the applications. Under virtually identical conditions with respect to temperature, humidity, light intensity and concentration, differences in reactions were found: sometimes damage and at other times no damage were found.

No striking differences were noted between longitudinal growth and the number of leaves on the treated and untreated plants, nor between the dry and fresh weight.

The following important points were noted:

1. A synergistic action of ozone with SO_2 was observed (possibly with other components). Therefore, it will be necessary to use a filter system so that purified air can be applied.

2. The plants seemed to be more sensitive to a specific ozone concentration if the greenhouse had been used for a SO_2 gas application the day before. Traces of SO_2 are released from the walls, the floor and the platforms for quite some time after an application. This can change the plant's sensitivity to O_3 and SO_2 for each location in the greenhouse. A greater number of gas application greenhouses is proposed to clearly separate the various gas application projects.

TABEL I

	Condities tijdens de begassing met PAN [1]					Het totale percentage schade van de bladeren [2]								De hoeveelheid licht [3]		
zaai-	begas.	conc.	temp.	R.V.	begas.	68853		69854		68206		69010		uren	gl. straling [9]	
datum [4]	datum [5]	(ppb)	°C	°/o [6]	tijd [7]	+	-	+	-	+	-	+	-	zon [8]	cal/cm^2	J/cm^2
3/5 - '73	12/6-'73	100	25 - 35	65 - 50	9^{00}-12^{00}	45	15	115	35	30	5	140	15	13,2	665	2788
3/5 - '73	12/6-'73	100	35	50 - 50	14^{00}-17^{00}	-	-	60	35	30	5	145	30	13,2	665	2788
3/5 - '73	13/6-'73	100	24 - 27	67 - 62	9^{00}-12^{00}	35	35	15	15	20	15	135	25	5,2	391	1640
3/5 - '73	13/6-'73	100	29 - 35	60 - 65	14^{00}-17^{00}	15	15	65	60	gs [10]	gs	165	55	5,2	391	1640
10/5 - '73	20/6-'73	100	25 - 28	68 - 68	8^{30}-11^{30}	10	gs	10	gs	gs	gs	10	5	7,2	474	1985
10/5 - '73	20/6-'73	100	33 - 27	60 - 72	14^{15}-16^{15}	gs	gs	gs	gs	gs	gs	20	15	7,2	474	1985
17/5 - '73	26/6-'73	100	28 - 33	65 - 66	9^{00}-12^{00}	5	gs	10	10	gs	gs	45	5	11,1	600	2513
17/5 - '73	26/6-'73	100	35 - 36	52 - 58	14^{00}-17^{00}	gs	gs	gs	gs	gs	gs	10	5	11,1	600	2513
17/5 - '73	27/6-'73	100	30 - 34	68 - 72	9^{00}-12^{00}	gs	gs	gs	gs	gs	gs	20	10	8,3	553	2316
17/5 - '73	27/6-'73	100	36 - 37	70 - 72	14^{00}-17^{00}	gs	gs	gs	gs	gs	gs	25	25	8,3	553	2316

Table I

Key:
1. Condition during PAN application
2. Total percentage damage of the leaves
3. Amount of light
4. Sowing date
5. Date of application
6. Relative humidity
7. Period of application
8. Hours of sunshine
9. gl. radiation
10. No damage

TABEL II

	Condities tijdens de begassing met PAN [1]					Het totale percentage schade van de bladeren [2]								De hoeveelheid licht [3]		
zaai-datum [4]	begas. datum [5]	conc. (ppb)	temp. °C	R.V. % [6]	begas. tijd [7]	68853		69854		68206		69010		uren [8]	gl. straling [9]	
						+	-	+	-	+	-	+	-		cal/cm²	J/cm²
3/5 - '73	14/6-'73	25	22 - 24	65 - 72	9^{30}-12^{30}	gs	gs	gs	gs	gs	gs	gs	gs	14,4	665	2788
10/5 - '73	18/6-'73	50	25 - 28	70 - 58	9^{00}-11^{00}	gs	gs	gs	gs	gs	gs	gs	gs	11,0	600	2513
10/5 - '73	18/6-'73	50	32 - 30	50 - 60	14^{00}-17^{00}	gs	gs	gs	gs	gs	gs	gs	gs	11,0	600	2513
10/5 - '73	19/6-'73	50	19 - 31	67 - 63	9^{00}-12^{00}	5	5	gs	gs	gs	gs	35	15	11,7	625	2619
10/5 - '73	19/6-'73	58	34 - 34	57 - 55	14^{00}-17^{00}	gs	gs	gs	gs	gs	gs	5	5	11,7	625	2619
10/5 - '73	21/6-'73	50	25 - 31	75 - 70	10^{00}-14^{00}	5	gs	gs	gs	-	-	10	gs	6,7	423	1774

Table II

Use the key for Table I.

TABEL III[a]

		Condities tijdens de begassing met PAN[1]					Beschadiging[2]			
Serie	poot-datum[3]	begas. datum[4]	conc. (ppB)	temp. ^oC	R.V. $^o/_o$	begas. tijd[5]	A	M	Dm	uren zon[6]
II	21/9-'73	24/9-'73	190	17 - 22	78 - 72	10^{00}-13^{00}	28,3	13,3	25,0	3,5
	21/9-'73	24/9-'73	190	21 - 24	67 - 67	13^{00}-16^{00}	18,3	6,7	13,0	0,9
	21/9-'73	26/9-'73	175	17 - 23	80 - 70	9^{00}-12^{00}	43,3	86,6	120,0	0,5
	21/9-'73	26/9-'73	175	25 - 23	70 - 75	13^{00}-16^{00}	37,5	66,7	73,3	-
	21/9-'73	28/9-'73	175	17 - 20	87 - 80	9^{00}-12^{00}	68,3	48,3	78,3	2,1
	21/9-'73	28/9-'73	175	22 - 25	75 - 65	13^{00}-16^{00}	38,3	48,3	106,5	-
	21/9-'73	25/9-'73	75	20 - 24	76 - 66	11^{00}-15^{00}	3,3	1,7	4,2	1,3
	21/9-'73	27/9-'73	75	22 - 25	80 - 75	11^{00}-15^{00}	gs[7]	gs	3,3	0,5
III	28/9-'73	1/10-'73	175	17 - 21	76 - 70	9^{00}-12^{00}	25	35	35	4,9
	28/9-'73	1/10-'73	175	22 - 22	74 - 74	13^{00}-16^{00}	45	170	100	1,1
	28/9-'73	1/10-'73	175	20 - 24	76 - 71	11^{00}-15^{00}	gs	gs	gs	2,2

Key:
1. Conditions during PAN gas application
2. Damage
3. Sowing date
4. Gas application date
5. Period of application
6. Hours of sunshine
7. No damage

/ Translator's note: Commas in the tabulated figures are equivalent to decimal points./

TABEL III^{b}

		Condities tijdens de begassing met PAN[1]					Beschadiging[2]			
Serie	poot-datum[3]	begas. datum[4]	conc. (ppb)	temp. °C	R.V. °/o	begas. tijd[5]	A	M	Dm	uren zon[6]
IV	4/10-'73	10/10-'73	175	15 - 20	82 - 72	$9^{00}-12^{00}$	5	1,7	5	0,4
	4/10-'73	10/10-'73	175	23 - 21	60 - 61	$13^{00}-16^{00}$	10	8,3	5	-
	4/10-'73	12/10-'73	175	17 - 19	74 - 62	$9^{00}-12^{00}$	11,7	6,7	5	3,2
	4/10-'73	12/10-'73	175	19 - 20	65 - 65	$13^{00}-16^{00}$	11,7	6,7	5	1,2
	4/10-'73	9/10-'73	175	21 - 21	64 - 64	$11^{00}-15^{00}$	gs[7]	gs	gs	-
	4/10-'73	11/10-'73	175	17 - 20	68 - 65	$11^{00}-15^{00}$	gs	gs	gs	-
V	11/10-'73	15/10-'73	175	14 - 20	74 - 68	$9^{00}-12^{00}$	8,3	-	8,3	-
	11/10-'73	15/10-'73	175	18 - 21	65 - 65	$13^{00}-16^{00}$	5,0	gs	13,3	-
	11/10-'73	17/10-'73	175	14 - 20	72 - 66	$9^{00}-12^{00}$	6,7	5	6,7	1,7
	11/10-'73	17/10-'73	175	20 - 20	65 - 65	$13^{00}-16^{00}$	3,3	gs	8,3	0,1
	11/10-'73	18/10-'73	175	14 - 19	70 - 64	$9^{00}-12^{00}$	gs	6,7	gs	1,2
	11/10-'73	19/10-'73	175	15 - 20	65 - 61	$9^{00}-12^{00}$	3,3	13,3	6,7	-
	11/10-'73	19/10-'73	175	17 - 20	65 - 65	$13^{00}-16^{00}$	gs	5	gs	-

Table IIIb

Use the key for Table IIIa

TABEL III^{c}

		Condities tijdens de begassing met PAN [1]					Beschadiging [2]			
Seria	poot-datum [3]	begas. datum [4]	conc. (ppb)	temp. ^{o}C	R.V. $^{o}/_{o}$	begas. tijd [5]	A	M	Dm	uren zon [6]
V	11/10-'73	16/10-'73	75	15 - 20	68 - 65	11^{00}-15^{00}	gs [7]	gs	1,7	-
VI	19/10-'73	22/10-'73	175	16 - 21	66 - 65	9^{00}-12^{00}	3,3	3,3	gs	0,1
	19/10-'73	22/10-'73	175	21 - 22	65 - 64	13^{00}-16^{00}	gs	1,7	1,7	-
	19/10-'73	25/10-'73	175	13 - 19	71 - 69	9^{00}-12^{00}	5	41,6	3,3	0,9
	19/10-'73	26/10-'73	175	13 - 18	75 - 74	9^{00}-12^{00}	1,7	8,3	68,3	3,9
	19/10-'73	23/10-'73	175	18 - 21	64 - 64	11^{00}-15^{00}	gs	gs	gs	-
VII	26/10-'73	29/10-'73	175	18 - 21	72 - 70	9^{00}-12^{00}	5	10	20	2,5
	26/10-'73	29/10-'73	175	19 - 20	77 - 78	13^{00}-16^{00}	15	13,3	58,3	-
	26/10-'73	31/10-'73	175	14 - 19	80 - 74	9^{00}-12^{00}	gs	gs	gs	1,7
	26/10-'73	2/11-'73	175	15 - 20	68 - 64	9^{00}-12^{00}	23,3	26,7	gs	-
	26/10-'73	2/11-'73	175	18 - 21	65 - 66	13^{00}-16^{00}	gs	gs	gs	-

Table IIIc

Use the key for Table IIIa

TABEL III^{d}

		Condities tijdens de begassing met PAN [1]					Beschadiging [2]			
Serie	poot-datum [a]	begas. datum [4]	conc. (ppb)	temp. °C	R.V. °/o	begas. tijd [5]	A	M	Dm	uren zon [6]
VII	26/10-'73	30/10-'73	75	21 - 19	75 - 73	11^{00}-15^{00}	gs [7]	gs	gs	-
	26/10-'73	1/11-'73	75	16 - 21	68 - 68	11^{00}-15^{00}	gs	gs	gs	-
VIII	1/11-'73	5/11-'73	175	18 - 22	71 - 66	9^{00}-12^{00}	96,6	100	145	-
	1/11-'73	5/11-'73	175	21 - 17	67 - 67	13^{00}-16^{00}	gs	3,3	gs	-
	1/11-'73	6/11-'73	175	14 - 21	72 - 65	9^{00}-12^{00}	10	11,6	26,6	0,3
	1/11-'73	6/11-'73	175	20 - 21	59 - 62	13^{00}-16^{00}	5	3,3	8,3	-

Table IIId
Use the key for Table IIIa.

TABEL IV^{a}

$^{o}/_{o}$ Droog/Vers gewicht[1] Ras: Amanda+[2]

Begast[3]		9^{00}-12^{00} uur[5]	13^{00}-16^{00} uur	11^{00}-15^{00} uur	
Concentratie[4]		175 ppb	175 ppb	75 ppb	Controle[6]
week	serie				
38	I	7,19 ± 0,17			7,01 ± 0,89
39	II	6,54 ± 0,20	6,90 ± 0,14	6,37 ± 0,10	6,10 ± 0,30
40	III	6,36 ±	7,28	6,63	6,32 ± 0,30
41	IV	6,06 ± 0,10	6,07 ± 0,28	6,14 ± 0,14	6,05 ± 0,33
42	V	7,94 ± 0,35	7,45 ± 0,10	7,53	7,90 ± 0,61
43	VI	7,81 ± 0,14	8,22	8,46	7,49 ± 0,35
44	VII	6,39 ± 0,14	6,06 ± 0,24	6,89 ± 0,87	5,89 ± 0,46
45	VIII	6,30 ± 0,48	6,06 ± 0,14		5,67 ± 0,20

Table IVa

Key:
1. % dry/fresh weight
2. variety
3. gas application
4. concentration
5. time
6. control group

/Translator's note: Commas in the tabulated figures are equivalent to decimal points./

TABEL IV^{b}

$^{o}/_{0}$ $^{Droog}/_{Vers}$ gewicht 1				Ras: Deci minor 2	
Begast 3		$9^{00}-12^{00}$ uur 5	$13^{00}-16^{00}$ uur	$11^{00}-15^{00}$ uur	
Concentratie 4		175 ppb	175 ppb	75 ppb	Controle 6
week	serie				
38	I	6,38 ± 0,32			6,58 ± 0,59
39	II	6,06 ± 0,17	6,15 ± 0,40	5,77 ± 0,10	5,58 ± 0,22
40	III	6,36 ±	6,32	6,00	5,82 ± 0,28
41	IV	5,49 ± 0,08	5,74 ± 0,33	5,68 ± 0,41	5,54 ± 0,24
42	V	7,41 ± 0,28	7,69 ± 0,36	7,40	7,13 ± 0,48
43	VI	7,33 ± 0,22	8,07	7,13	7,12 ± 0,32
44	VII	5,69 ± 0,22	5,89 ± 0,47	5,49 ± 0,0	5,67 ± 0,39
45	VIII	6,12 ± 0,41	5,55 ± 0,10		5,27 ± 0,22

Table IVb

Use key for Table IVa.

TABEL IV^{c}

°/o Droog/Vers gewicht [1] Ras: Miranda [2]

Begast [3]		9^{00}-12^{00} uur [5]	13^{00}-16^{00} uur	11^{00}-15^{00} uur	
Concentratie [4]		175 ppb	175 ppb	75 ppb	Controle [6]
week	serie				
39	II	6,60 ± 0,98	6,02 ± 0,17	5,90 ± 0,17	5,68 ± 0,48
40	III	6,49	6,28	5,80	5,95 ± 0,36
41	IV	6,28 ± 0,79	5,94 ± 0,37	5,54 ± 0,14	5,92 ± 0,62
42	V	7,05 ± 0,57	7,33 ± 0,36	6,34 ±	6,50 ± 0,69
43	VI	7,50 ± 0,22	7,69	6,69	7,48 ± 0,47
44	VII	5,59 ± 0,14	5,57 ± 0,44		5,68 ± 0,24
45	VIII	6,05 ± 0,40	5,47 ± 0,0		5,14 ± 0,10

Table IVc

Use key for Table IVa.

APPENDIX IIIB

1973 Review of the Biological Tests for the "Smog Project"

Research Institute for Plant Diseases, Wageningen

(located at the Experimental Station for Greenhouse Cultures
at Naaldwijk, the "Proefstation voor de Groente - en Fruitteelt onder Glas")

1. Introduction

The Institute contributed to the project the detection and measurement of various air pollution components with the aid of biological components.

2. Set Up of the Experiment

Based on the experience of past years with artificial gas applications at the Institute, it was decided to start a detection system of a number of small test fields on which indicator plants would be grown. In addition it was decided to install some small greenhouses, half of them supplied with a filter system to purify the air, at various locations in the region.

2.1. Test Fields

These test fields were prepared last winter. The following list shows the locations of the various fields.

1. Naaldwijk Experimental Station for Greenhouse Cultures, located at TNO-RIV
2. Maasdijk, Tuindersweg
3. Maasland, village grounds; also the location of a mobile unit

4. Schipluiden, within the built-up area
5. Delft, grounds of the TNO, next to test location I.G. TNO
6. Vlaardingen, Florislaan, test point R.I.V.
7. Barendrecht, Stationbuurt
8. Smitshoek (center of village)
9. Poortugaal, grounds of the Delta hospital
10. Heenvliet (center of village)
11. Oostvoorne (in the center, village grounds)
12. Rockanje, near Volkshogeschool
13. Hellevoetsluis, grounds of Rijkswaterstaat, test point R.I.V.
14. Vierpolders test garden.

All these fields were installed with so-called cultivation containers. These containers have filter candles which automatically supply water. This system has the additional advantage that we can start with a homogeneous soil mixture, thus eliminating a cause of possible differences in growth.

2.1.1. Indicators

a. Spinach

Selected because damage is observed frequently in the past by growers in the region Oostvoorne-Rockanje. Indicator for $SO_2 + O_3$. Two varieties were used, Subito and Dynamo.

b. Luzerne

Sensitive to SO_2, and according to American literature, also for ozone. Variety "Du Puits"

c. Petunia

Known as an indicator for ethylene (C_2H_4); for lower concentrations, 12 hours at 0.1 dpm, a smaller flower diameter is observed; longer exposure suppresses flowering altogether. Cultivar: "White Satin".

d. Small Stinging Nettle

Indicator for Peroxyacetylnitrate (PAN). Well known is the clear band shaped damage extending over the entire width of the leaf.

e. Freesia

Indicator for hydrogen fluoride (HF). Cultivar: "Golden Yellow".

f. Gladiolus

Also for HF. Cultivar "Snow Princess".

g. Tulip

Sensitive to SO_2 and HF. Cultivar: "Blue Parrot".

h. Tobacco

Indicator for ozone. Damage pattern: big and small spots on the leaves. Variety: Bel W3.

i. Street grass

Indicator for Peroxyacetylnitrate.

j. Yellow Mustard

According to the literature, an indicator for PAN and NO_2. Tried out in the indicator system. No results obtained.

k. African Marigold: Tagetes (Er.)

Also a try out. Indicator for ethylene. The entire season, no damage symptoms observed.

l. Snijbiet (a variety of beets)

According to the literature, sensitive to PAN and SO_2. Try out.

m. Rhubarb

Tried out on a few fields as an indicator for NO_2.

n. Kale

This indicator used in Germany is insensitive to air pollution, but it is known that deposits of chemicals like cobalt, sulphur and fluor form on the leaves. With leaf analysis these deposits can be measured.

2.1.2. Results Obtained with the Plant Indicators on the Test Fields

For spinach it was striking that the greatest damage occurred in May at the southern edge of the Nieuwe Waterweg. This was similar to the the observations of the previous years when the plants were already damaged in the middle of April and the beginning of May, close to the harvest time. This damage did not coincide with storm or night frost. It was not possible anymore to trace whether ozone or SO_2 was the cause. The Petunia showed remarkable leaf burns: a gloss both at the front and back side of the leaf, changing most of the time into burned spots. Probably PAN symptoms.

The small stinging nettle was clearly damaged at the north side of the Nieuwe Waterweg in the weeks September 10-16 and 17-23. This conformed to the measurements of the Central Laboratory T.N.O. at Delft, which showed the presence of peroxylacetylnitrate (PAN) in Delft.

Street grass showed damage too in the same two weeks.

Gladiolus and freesia clearly showed the HF emission from Vlaardingen.

Tobacco showed serious leaf damage with the typical ozone damage pattern in the period June 18-24, August 27-September 2 and Sept. 10-26.

2.2. Experimental Greenhouses

a. Naaldwijk test station

   SO_2 filter

   oxydantia filter

   ethylene filter

   control with axial ventilator

   control with centrifugal ventilator

   control with centrifugal ventilator + dust filter

   control with axial ventilator + dust filter

b. Den Haag, Zuiderpark

   control

   oxydantia filter

c. Delft, T.N.O. Zuidpolder

   control

   oxydantia filter

d. Vlaardingen, Florislaan

   control

   oxydantia filter

e. Hellevoetsluis, grounds of Rijkswaterstaat (National Water Board)

   control

   oxydantia filter

f. Maasdijk, Tuindersweg

At this location there are four greenhouses, of which two are outside. The other two are inside a heated greenhouse from the Beekenkamp Company. These two are used for ethylene test for tomatoes.

2.2.2. Indicators in the Greenhouses

Already described are the indicators that were also used for the test fields: tulip, petunia, small stinging nettle, street grass, luzerne, tobacco, kale, gladiolus, freesia. An indicator for ethylene used only in the greenhouse was the tomato. This plant was selected not only for its temporary curvatures observed in the presence of the gas, but also so as to study the indirect damage (diminished growth and a smaller harvest) caused by the frequent exposure to very low concentrations (0.1 dpm) of ethylene. The great economic importance of the tomato for this region was another factor that favored its selection.

2.2.3. The Test on the Climate in the Greenhouses and the Use of Filters

The purpose of the preliminary research in 1973 was the preparation of the greenhouses in the Nieuwe Waterweg region for the demonstration of air pollution according to a new method.

Half of the greenhouses are connected to the outside atmosphere by way of an axial ventilator; the other greenhouses are provided with a filter which allows air to be sucked in with the aid of a centrifugal ventilator, which blows the purified air in the greenhouses. Three types of filters were available (carbon filters, Norit): Oxydantia filter, SO_2 filter, and ethylene filter.

Already in the past great differences in growth were observed between plants cultivated in greenhouses with and without filters. Therefore a test was initiated to see whether these differences were caused by:

a. Differences in climate caused partially by the construction of the greenhouses;

b. The action of the carbon filter, the filtering away of air polluting components in the greenhouse.

Advice was obtained from the Institute of Health Technology T.N.O., Delft, the test station of the "Groente - en Fruitteelt onder Glas", Naaldwijk, and the "Stichting Technische en Fysische Dienst voor de Landbouw," Wageningen.

The tests were performed at the greenhouses of the Naaldwijk test station. The seven greenhouses that were used in the test were:

- two control greenhouses with an axial ventilator
- one control greenhouse with a centrifugal ventilator + dust filter
- one control greenhouse with a centrifugal ventilator
- one oxydantia filtered greenhouse with a centrifugal ventilator
- one ethylene filtered greenhouse with a centrifugal ventilator
- one sulphur dioxide filtered greenhouse with a centrifugal ventilator.

All measurements were performed in these greenhouses without plants.

a.1. The measured climate factors are:

1. Temperature
    a. Temperature differences between the greenhouses
    b. Temperature differences in the greenhouse for different locations
2. The air ventilation and distribution in the greenhouses.

a.1.1. The temperature is measured with a thermo couple of copper constantane. This information was registered by a recorder.

a.1.2. The air ventilation and distribution is measured with anemometers:

a. A wind velocity monitor for the ventilation (in m/min)

b. Heat coil anemometers for the distribution (in cm/sec)

b.1. The measured air pollution components are:

b.1.1. SO_2

b.1.2. O_3

b.1.3. NO_2.

The various air pollution components are measured by letting outside air flow through a reagent and analyzing the sample with a spectrophotometer.

2.2.4. Results of the Greenhouse Tests

The results of the preliminary tests of the air ventilation will be given first.

3.a. Climate tests

Climate tests showed the great influence of the amount of air (per time unit) that flows through the greenhouse upon the temperature.

An effort was made to equalize the air velocity to 7 m^3/min for all greenhouses. This is very well possible in the greenhouses with a centrifugal ventilator:

- in greenhouses with a filter by varying the thickness of the filter layer;
- in greenhouses without a filter by varying a valve's position from horizontal (open) to vertical (closed).

There is no way to measure or control the air velocity in the greenhouses with an axial ventilator (used for the control greenhouses).

These greenhouses were always lower in temperature, according to the temperature measurements, so it must be assumed that the air velocity is higher than ± 4 m^3/min, the original assumption, and also higher than 7 m^3/min, the air velocity in the greenhouses with a centrifugal ventilator.

The air distribution in the greenhouses is also much more irregular and fluctuates more than that in the greenhouses with a centrifugal ventilator. This probably also causes part of the difference in growth of the plants, which is also observed in the control greenhouses supplied with [illegible] axial ventilator.

Mainly because of the lower temperatures in greenhouses with an axial ventilator and the fact that the air velocity of these ventilators cannot be measured, the decision was made to use exclusively "relative comparable" centrifugal ventilators.

3.b. The Action of the Carbon Filters

SO_2: The three applied carbon filters showed the following results:

- the SO_2 filter let in ± 4% of the SO_2 air contents from the filter-circulated outside air during the spring period.
- The oxydantia filter let in ± 13%, and the ethylene filter let in ± 21% of the outside air SO_2 contents. The results of the measurements are given in Table I.

O_3: In none of the greenhouses with a carbon filter was the presence of ozone detected. In the control greenhouses ozone was found, but the concentration was lower than that of the outside air.

NO_2: Almost no NO_2 is let through by the oxydantia filter. The sulphur dioxide allowed less than half of the outside air NO_2 concentration to pass through. The ethylene filter allowed about half of the outside air NO_2 through. Meetings with Norit N.V. research laboratories are initiated in order to improve the specificity of the filters.

3.c. Results with the Plant Indicators Grown in the Test Greenhouses

Tulip

Planted in January at the following locations: Naaldwijk, Delft, Maasdijk, Vlaardingen. Rather serious damage occurred between March 26 and April 8.

Tobacco

Remarkable are the extreme differences in weight measurements that can occur. It seems useful to study this phenomenon more. However, it is necessary that the climates in the filter greenhouse and the control greenhouse are equal as much as possible.

Tomato

For the tomato the comments made for tobacco are applicable. However, because of the great economic importance of the tomato, the situation is more serious in this case.

Short Summary of the Results Obtained

At various periods during the growth season of the indicator plants for air pollution components, damage was observed, caused by the action of ozone and peroxyacetylnitrate (PAN); for hydrogen fluoride a gradual development of the damage during the growth season was observed.

In greenhouses supplied with an activated carbon filter for removal of the air polluting components, the plants grew better than in unfiltered greenhouses. It is not clear yet that this is caused by the removal of the air polluting components because careful measurements showed some differences in temperature between the greenhouses with and without filters. It was investigated whether it was possible to obtain an equal climate in both types of greenhouses, which would exclude growth differences caused by small differences in climate. It will be possible in this way to find the causes of possible growth differences.

In the period from March 26 to April 8, leaf burning of the tulip plants was observed in the unfiltered greenhouses at four of the six locations; the damage looked like sulphur dioxide damage.

The detailed results of the experiment are kept at the Research Institute for Plant Diseases and can be obtained upon request.

Tabel I

Uitkomsten van de SO_2 metingen in de verschillende kasjes te Naaldwijk.

Kasje I = SO_2 filter

		SO_2 gehalte 2	
		in kasje 5	in buitenlucht 4
Meting: 3	1	2 ug/m^3	95 ug/m^3
	2	4	86
	3	n.a.	270
	4	4	115
	5	2	86
	6	4	118
	7	n.a.	145
	8	11	208
	9	13	336
	10	6	130

Kasje II = oxydantia filter

		SO_2 gehalte	
		in kasje	in buitenlucht
meting:	1	30 ug/m^3	101 ug/m^3
	2	21	152
	3	14	122
	4	11	108
	5	16	154
	6	2	107
	7	14	72
	8	5	76
	9	15	54
	10	13	87

Table I

Results of the SO_2 Measurements in Various Greenhouses at Naaldwijk

Key: 1. Greenhouse 3. Measurement 5. In greenhouse
2. Concentration 4. In outside atmosphere

Kasje III = ethyleen filter — SO_2 gehalte

	in kasje	in buitenlucht
meting: 1	25 ug/m^3	143 ug/m^3
2	23	80
3	21	95
4	13	75
5	20	81
6	20	84
7	10	89
8	14	79
9	12	104
10	16	96

Kasje IV = controle + stoffilter[1] — SO_2 gehalte

	in kasje	in buitenlucht
meting: 1	77 ug/m^3	102 ug/m^3
2	69	104
3	73	118
4	86	142
5	94	145
6	60	89
7	157	219
8	112	152
9	135	194
10	103	152

Key: 1. Greenhouse IV = controle + dust filter
(See page 90 for key.)

Kasje V = zonder filter SO_2 gehalte

		in kasje	in buitenlucht
meting:	1	39 ug/m^3	45 ug/m^3
	2	48	46
	3	55	55
	4	54	55
	5	106	121
	6	49	53
	7	42	57
	8	25	40
	9	68	104
	10	78	118

Key: 1. Greenhouse V = without filter
(See page 90 for key.)

APPENDIX IV

Overview of the Air Pollution Measurements with a Mobile Unit by KvW/DCMR for the Experimental Project South-Holland

J. van Straaten

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2. Description of Measurements

2.1. The Measurements Network

The selected region included the following counties after consultation with the members of the Experimental Project South Holland: Vlaardingen, Schiedam and Rotterdam, including Hoogvliet and Pernis. With the aid of the current coordinate system of the Topographic Service in Delft, a two km latice is projected on the sampling region. The possible sampling locations are found at the intersections of the coordinates. Of course, not all locations are suitable because they might be located at a river site, a meadow or an inaccessible region. Finally eighteen locations were selected; some of these had to be slightly relocated because of local conditions. Originally a location at Nieuwelaan, in the center of Vlaardingen, was selected. However, almost immediately a location at the Floreslaan was chosen instead. Reasons were the sanitation program of the Nieuwelaan district and the fact that at the Floreslaan already a stationary sampling station of the Project was located. This makes it possible to compare the results of the stationary station and the mobile unit.

The eighteen sampling points are described in Table 1 in the following order: an internal serial number, the R.I.V. code used until January 1, 1974, which was replaced afterwards by a code containing the coordinates. The coordinates are written next to each other in the next column of the Table and are followed by the address. The next two columns contain wind sectors that will be explained later on in the report. The location of the sampling points is shown in Fig. 1, which shows also the "routes." The last column indicates the way in which sampling locations in the routes are combined.

2.2. The Sampling Program

The samples were taken by the mobile unit in the period from October 1975 to December 1975, workdays between 8 a.m. and 5 p.m. Sampling time was mostly half an hour, and it took approximately half an hour to drive to the next location. Each day six locations on the same route were sampled. The remainder of the day was used for calculations and servicing the car and instruments. Although ideally the sampling should take place all day, reality shows that this is not possible (see frequencies in Fig. 2). At the most, each of the locations can be visited 52 times a year, but in reality this theoretical number of 936 samples could not be reached. In order to visit these 18 sampling points, a scheme was devised that indicated three routes and the starting points of the routes. The routes were always driven counter-clockwise. The order of the routes and the starting points were selected at random. Not included in the scheme were the weekdays at which a certain route with a certain starting point was driven, because this is very much dependent on the circumstances related to the Institute. The final distribution of the sampling of each route over the working days in each quarter is shown in Table 2 and Fig. 4.

2.3. Instrumentation

As a rule at each location of the project South Holland, nitrogen monoxide (NO), nitrogen dioxide (NO_2), carbon monoxide (CO) and ozone (O_3)

are measured. The nitrogen oxides are measured in separate monitors (Enraf-Nonius). Besides a few incidental troubles these instruments performed pretty well. The correctness of the measurements is not discussed here because the instruments were gauged by the R.I.V. The CO monitor (Maihak) turned out to be more sensitive to moisture, mechanical vibrations and radio frequencies (mobilophone). Because of the low CO level with respect to the full scale of the instrument (1-3 for a 100 ppm full scale), except for traffic measurements, the above mentioned factor caused a lot of trouble, such as too high or even negative readings. These readings are therefore, with the exception of those of the fourth quarter of 1973, questionable and are not used in this report.

The O_3 monitor (Euroglas) showed sensitivity for radio frequencies; moreover, the instrument turned out to be not very suitable for mounting in a car because of the adjustment of a potential equilibrium at the electrodes. Nonetheless, the measured values give a good indication of the ozone concentrations because a stabilization period was adhered to. Each of the monitors has its own sucking mechanism. This sucks the outside air in through a central system which has a vacuum of approximately 0.8 cm water column. All pipes are made of teflon and are connected by glass couplers. The data from the monitors are fed into the data logger (Enraf-Nonius), processed and recorded, marked with datum and time, by an ASR (teletype) on paper and punch tape.

2.4. Conditions During Measurements

As described before, the order of the measurements is previously scheduled. However, there is room left to choose the working day and the hour, depending on the circumstances. To test the desired randomness of the sampling, counts of samples under various conditions were performed.

In Fig. 2 the sample frequency versus the time of the day is plotted. This shows that almost all samples are taken between 9 a.m. and 5 p.m. The temperature for each day follows a definite pattern, and this temperature might influence the NO/NO_2 ratio. Therefore the relation between the sample frequency and the outside temperature is investigated. Figure 2 shows this relationship. Table 2 shows the sample frequency per route and per weekday for the five quarters and the year 1973. The totals per weekday of 1973 are shown graphically in Fig. 4. Although Thursday seems to be slightly favored, the distribution of the samples over the working days seems reasonable.

Finally Fig. 5 shows the sample frequency for each wind direction. For an ideal test this distribution should be equal to the wind direction frequency (wind direction x time period)/total time, which is recorded by the National Meteorological Institute. These two distributions are roughly equal. The few discrepancies can be explained by the fact that the wind frequencies are measured over 24 hours a day, while the distribution of samples over the wind directions refers to time periods between 8 a.m. and 5 p.m., and this only for working days.

3. Results of the Measurements (First Processing)

The original results of the measurements, obtained by the mobile unit in the period 1972 to December 1973, are given in Appendix I. The order is chronological for each location. For each sample the Table includes: the location, day of the week, datum, sampling time, measured values of NO, NO_2 and O_3 in $\mu g/m^3$ and CO in ppm, the ratio NO/NO_2, the wind direction and the temperature. From these data the following are computed: average, standard deviation, divergence and number of samples per component. These are given in Appendix II for NO and NO_2. For the fourth quarter of 1973 these quantities are also computed for CO because only for this period did the measurements obtained seem to be trustable. The appendices are found in a volume separate from this report.

3.1. Iso-lines

The 1973 averages are shown graphically with the aid of iso-lines for each location. This method shows the geographical location of certain concentrations. The iso-lines between the sampling points are obtained by linear interpolation and give a fair impression of the situation in the sampling region. The divergence of the averages is not accounted for. It must be noted that the drawing of iso-lines with the available data is rather arbitrary. The "NO-1973" map shows two nucleus regions, one above Vlaardingen and a bigger one above the city Rotterdam. However, the line pattern above Rotterdam cannot be considered homogeneous, because the sampling locations near 26 Rochussenstraat, near 18 Lombardkade and near 43 Slinge, are traffic locations. One would expect a relatively high NO concentration and the data confirm this.

The "NO_2-1973" map in Fig. 7 shows a similar distribution of the averages per location. Here again a two-nucleus pattern is observed. In the biggest nucleus two separate peaks can be observed. This confirms pretty well Larsen's findings. It becomes clear that for both NO and NO_2 an in-between region exists which accidentally (?) does not contain as many sampling locations. These regions (Schiedam-Rhoon-Poortugaal) have to be more emphasized in the future.

The final iso-line map is the "NO/NO_2-1973" map in Fig. 8. It looks very much like the NO and NO_2 maps. Because of the temperature dependence explained later on in this report, not much can be said about the yearly average. Of course this could be investigated for a quarter or a season, but not quite enough information is available for this.

3.2. Comparison of the Quarter and the Season Levels with the Yearly Average

The averages per location and for the entire region per quarter and combination of quarters are given in Table 3 for NO, in Table 4 for NO_2 and in Table 5 for NO/NO_2. Figure 9 shows a selection of these data graphically. This figure shows a clear difference between the colder and warmer periods of the year. The NO levels fluctuate more than the NO_2 levels. The NO/NO_2 ratio is not only influenced by the temperature but also by the NO level. The regression analysis shows a generally clear relation between NO level and the NO/NO_2 ratio. The Figs. 10-17 show these relations. It is remarkable that all correlations, with the exception of the second quarter of 1973 and the second and third quarters of 1973 taken together, are above the significance level.

4. Results of Measurements (Direction of Wind)

To obtain results suitable for the project South Holland, the available data are processed according meteo-conditions. The original sample values are for this purpose selected according to direction of wind and temperature.

This gives a better insight into:

a. the effect of the wind direction on NO_x levels, and

b. the effect of the wind direction on the NO/NO_2 ratio.

The most visible effects will be discussed first. Later, other effects will be added as well. The meteo information, the wind direction and temperature, are indicated for each sample in the tables of Appendix I.

4.1. Compass Card

For a great number of samples per location the compass card is very suitable in defining the direction of the sources. For two locations, Sweelincksingel at the right bank of the Maas River and Schulpweg at the left bank, two compass cards are made for both NO and NO_2. To enable the plotting of the small number of samples at each location, the samples are partitioned into wind sectors of 30°. Even then some sectors are empty or contain only one sample. Figures 18 and 19 show the NO and NO_2 compass cards. In each sector the number of samples per component, from which the average is computed, is indicated. At the sampling location Schulpweg the sector 150°-270° attracts attention. In this direction a busy highway is located: de Groenekruisweg. For the location Sweelincksingel the highest averages are in the direction of the industrial region between Pernis and Oude Maas. In both figures a NO peak is observed, but in both cases this is only one sample. The results suggest that another approach would be more suitable because of the small number of available samples.

4.2. Sectors

An approach that seems more promising is the partition of the compass card into some previously selected sectors. Therefore, for each sampling location the following sectors are defined:

- the "I" sector, which contains the entire industrial region
- the "K" sector, which contains the greenhouse region
- the "R" sector, which contains the rest of the sampled region.

The choice of these sectors is arbitrary with respect to size and direction, of course. Table 1 gives these sectors for each location. This partition according to source regions ("I" and "K") does not resemble the well known rule of thumb which attributes 1/3 of the total air pollution to each of the sources, Industry, Traffic and Heating of Homes. In both partitions industry is found. In the partition that is used for this report an "I" region, in which the industry is located, is used. One would expect, in line with the rule of thumb, a contribution of 1/3 for "I".

4.3. Selection of the Sectors "I", "K" and "R" from the Complete Sampling Region

To make it possible to find the relative contributions of "I", "K", and "R" for each sampling location a selection of the measured data is made based on the previously selected wind direction sectors. For all eighteen locations three groups were obtained this way. Groups of the same name were joined to obtain "I", "K" and "R" groups for the entire region in this way, notwithstanding the differences in size and direction of the individual sectors. At most locations "I" and "K" are separated, but at the sampling point at Hoogvliet "K" and "I" partially overlapped. Therefore these samples are not used in the computations. The results of this computation can, so to speak, be projected on the computed center of gravity of the sampling region, the "Z" in Table 1.

4.3.1. Computation of the Relative Contributions of "I", "K" and "R" in the Entire Region

To find the relative contributions of the various sectors, the following computations are performed:

For each sector the average, the divergence and the number of samples are computed for the four quarters of 1973. The divergence (r) is computed for $p = 0.05$ according to the equation

$$r = \pm \frac{1.96 \times \text{s.d.}}{\sqrt{n}}$$

However, in these computations the divergences are not used further on. The relative contribution (RB) for NO and NO_2 of a sector is computed according to

$$\frac{n_{s1} \cdot \bar{x}_{s1}}{(n_{s1} \cdot \bar{x}_{s1})+(n_{s2} \cdot \bar{x}_{s2})+(n_{s3} \cdot \bar{x}_{s3})} \times 100\% = RB$$

with n_s = number of samples in a sector

$\bar{x}_s$ = average level in sectors.

The averages, divergences and relative contribution percentages of NO and NO_2 for each quarter and the entire year 1973 are given in Table 6.

The relative contributions can be shown graphically like Figs. 20 and 21. These cumulative graphs show that a difference exists between the summer half year (second and third quarters) and the winter half year (first and fourth quarters). The same differentiation can be made for the average levels per sector and for the sectors together. Chapter 5 elaborates on this issue. Altogether the observed relative contribution of "I" matches quite well the rule of thumb percentage (24% versus 33%), with respect to the entire sampling region.

However, the above sketched method gives a very rough assessment. To obtain a closer assessment the following have to be considered:

a. Reliability intervals (for example, $p = 0.05$)

b. Overlapping "K" and "I" sectors at Hoogvliet cause the samples obtained at these sectors not to be included in the computations. This might have distorted the overall view.

c. For the sampling points in the east region, the sector "I" and "K" contain some contributions of "R" because a large part of town is included in the computations.

Therefore a limitation on the sampling region has to be made that will allow the picture to be as realistic as possible. Three Vlaardingen sampling locations are selected for a precise contribution computation.

d. The number of samples per sector is relatively low. Only for a very large number of samples will the ratio of the number of samples in a sector (n_s) to the total number of samples (n_t) be equal to the wind frequency in that sector (f_s). In this case the following inequality holds:

$$\frac{n_s}{n_t} \neq \frac{f_s}{f_t} = f_s \qquad (f_t = 1).$$

In the following computations the sector averages ($\overline{x}_s$) are coupled to the wind frequency in that sector (f_s), which makes the results independent from the number of samples.

e. No consideration is given to a pre-load factor. Therefore, in addition to a computation with the pre-load equal to zero ($V = 0\ \mu g/m^3$), computations in which the pre-load has a certain value will also be made.

4.4. Computations of the Relative Contributions of "I", "K" and "R" for NO and NO_2 in Vlaardingen

For each location or sub-region the sector averages ($\overline{x}_s$), the standard deviation (s_s), the number of samples (n_s) and the divergence (r_s) are computed for each sector. In addition for each sector the wind frequency (f_s) is obtained from the information of the National Meteorological Institute (airport Rotterdam).

The wind frequency is the resultant of the wind direction and the duration of this wind direction. However, no partition into wind velocity classes is made.

The relative contribution (RB) of sector 1 to the total level can be computed according to

$$RB = \frac{(f_{s1} \cdot \bar{x}_{s1})}{(f_{s1} \cdot \bar{x}_{s1}) + (f_{s2} \cdot \bar{x}_{s2}) + (f_{s3} \cdot \bar{x}_{s3})} \times 100\%$$

To obtain the maximal contribution ($p = 0.05$) of a sector, the upper limit of that sector and the lower limit of the other sectors can be used in the formula above:

$$RB_{(max)} = \frac{f_{s1} \cdot (\bar{x}_{s1} + r_{s1})}{f_{s1} \cdot (\bar{x}_{s1} + r_{s1}) + f_{s2} \cdot (\bar{x}_{s2} - r_{s2}) + f_{s3} \cdot (\bar{x}_{s3} - r_{s3})} \times 100\%$$

The same for the minimal contribution:

$$RB_{(min)} = \frac{f_{s1} \cdot (\bar{x}_{s1} - r_{s1})}{f_{s1} \cdot (\bar{x}_{s1} - r_{s1}) + f_{s2} \cdot (\bar{x}_{s2} + r_{s2}) + f_{s3} \cdot (\bar{x}_{s3} + r_{s3})} \times 100\%$$

Both formulas have to be computed three times for each sampling point this way.

Only for the assessment of the averages, which has a divergence of zero, the sum of the contribution percentage is 100%. In addition to Table 7, these results are also shown in diagrams with triangle coordinates (Figs. 22-31). These show a polygon limited by the upper and lower **limits** of the three contributions. Thus, inside these lines the true mutual relationships can be found.

In addition the intersection of the three lines that represent the various assessments is indicated inside this region. This all assumes the pre-load to be equal to zero. If we assume that the pre-load is equal in all wind directions (v $\mu g/m^3$), a computation similar to the one above can be made with v=0 and the same formulas if "$\overline{x}_s$-v" is used instead of "$\overline{x}_s$".

In the graph a shift, with respect to the v=0 figure, will occur in the direction of the 0% line of the sector with the lowest sector average. This shift can only occur between two extremes:

a. one of the sectors has a 100% contribution, or

b. the contribution of both sectors is equal.

This can be shown graphically: it is sufficient to indicate the shift of the assessment of the contribution because the standard deviation, and therefore the divergence of a group of numbers too, does not change when a constant is added or subtracted for all numbers. Therefore, the original region outside which the assessment of the contribution ratio cannot extend, will not change in form or surface area. In computing the assessments for various pre-loads, a line is found that interconnects the obtained assessment. When the shifting polygon touches a nul % line, the maximal pre-load has been reached.

The composition of the contribution for the maximal pre-load can be read from the triangular diagram at the intersection of the line along which the assessment of the averages shifts and the line that represents the difference in the assessment at V+0 and the distance from the intersection line to the null % line. In two of the triangular diagrams these lines are drawn.

4.5. Comparison of the Relative Contributions of "I", "K" and "R" in Vlaardingen and the Entire Sampling Region for NO and NO_2

The relative contributions of NO and NO_2 for the three Vlaardingen sampling locations and for these locations together are computed according to the methods given above for the year 1973. They are given in Table 7. For each sampling location in the table the contribution percentages of the three sectors for a pre-load of 0 $\mu g/m^3$ is given. Next to these the lower and upper limits for $p=0.05$ are given.

These percentages are quite different from the percentages found for the entire region. In Vlaardingen the relative NO contribution varies between 42% and 65%; in the entire region 52% is found. "K" in Vlaardingen is between 8% and 15%; 10% for the entire region; "R" is 27%-47% and 66% for the entire region. For NO_2 a similar shift is observed. "I" is 46%-64% in Vlaardingen, 26% in the entire region. "K" is 8%-11% and 11%; "R" is 28%-44% and 64%. Comparison of these percentages is not really allowed (see 4.3.1.) although a certain trend becomes visible that way. In Vlaardingen, which is located closer to the industrial region in sector "I" than the entire sampling region (projected on the center of gravity Z), a relatively greater contribution both for NO and NO_2 is found. Remarkably enough the 1973 average NO levels in Vlaardingen are lower than the average levels in the entire region: In Vlaardingen between 42 and 49 $\mu g/m^3$, in the entire region, 52 $\mu g/m^3$. For NO_2 in 1973 43-56 $\mu g/m^3$ and 52 $\mu g/m^3$ are found, respectively.

This might be caused by a number of factors; one could think of the following:

1. The NO emission of the industry goes partly to Vlaardingen.
2. The traffic intensity in Vlaardingen is lower than the remainder of the sampling region, and of course much lower than at sampling locations near highways.
3. A confirmation of Larsen's theory that a relation exists between the number of inhabitants of a city and the NO_2 level in that city; NO_2 is considered here as a derivative of NO.

From the computations performed on the available data it cannot be deducted which factor might be the right one. However, there are indications that the traffic contribution to the NO level may not be underestimated (see Fig. 6).

5. Results of Measurements (Time of the Day and Temperature)

5.1. Influence of the Time and the Temperature During the Measurements on the Measurement Results

While sorting the data, attention is also paid to the time of the day and the temperature during the measurements.

If the middle of the sampling period occurred between 8 a.m. - 9 a.m., the observations were classified in group 9, etc.

The average hour temperature in that period was looked up in the monthly meteo overview from the airport at Rotterdam. These were partitioned in ten classes of 3°C each (0-2, 3-5, ..., 27-29°C). For this investigation the data of all locations for the quarters were used. Furthermore, combinations of quarters in summer and winter half years are made. In this way for each quarter and combination of quarters, two frequency partitions could be made: one for the number of samples in each time period, and one for the number of samples in each temperature class (Figs. 2 and 3). These show that before 8 a.m. and after 5 p.m. no samples are taken. Therefore, a part of the morning peak and the entire evening peak are not measured. The number of samples in the periods 9 and 17 is smaller than in the other periods, and the results are therefore statistically less reliable.

5.2. Daily Chart

The average daily charts of the NO, NO_2 and NO/NO_2 values obtained from the data are drawn again for each quarter and for combinations of quarters (Figs. 32-39).

Remarks:

a. In the morning the levels are on the average higher than in the afternoon.

b. The NO levels fluctuate more than the NO_2 levels.

c. In the 1st and 4th quarter the levels are higher than in the 2nd and 3rd quarters.

d. In the 1st and 4th quarters the NO level is higher than the NO_2 level ($NO/NO_2 > 1$); in the 2nd and 3rd quarters NO/NO_2 is less than 1.

e. The NO/NO_2 ratio is minimal around noon (11 a.m.-2 p.m.).

f. This minimum appears earlier in the 2nd and 3rd quarters than in the 1st and 4th quarters. This indicates a temperature effect (see Fig. 40).

5.3. Temperature Chart

In the same way the average daily temperatures of NO, NO_2 and NO/NO_2 are charted (Figs. 41-48).

Immediately visible are the following:

a. The NO values rapidly decrease until approximately 15°C for increasing temperature ($-7\ \mu g/m^3/°C$) (Fig. 48) and stay approximately constant afterwards (less than $0.5\ \mu/m^3/°C$).

b. The NO values increase for higher temperatures above 15°C after a decrease between 1-4°C.

c. The NO/NO_2 ratio decreases for increasing temperatures; between 10-13°C the NO/NO_2 ratio is approximately 1.

d. For the same temperature the NO level is higher in the winter than in the summer quarters, and the NO_2 level in the winter quarters is lower than in the summer quarters for the same temperature.

e. The preceding point becomes even more clear in the NO/NO_2 charts. The NO/NO_2 ratio is for equal temperatures higher in the winter quarters than in the summer quarters (Figs. 49-57).

f. The fluctuations of NO are larger than those of NO_2.

g. The NO_2 level only increases when the NO level is almost constant (from approximately 15°C) (see Fig. 48).

The above leads to the conclusion that a relation between the temperature and the NO level, and between the temperature and the NO/NO_2 ratio exists.

The character of this relation has to be investigated more closely; a further selection of other meteo conditions might also influence this relation (wind direction and velocity, etc.). However, the sampling size does not allow a further selection.

6. Regression and Correlation Coefficients

There exists a visible similarity between the temperature charts of the NO level and the NO/NO_2 ratio (Figs. 48 and 57). To check this possible relation the correlation coefficient and the linear and second regression line are computed (see Figs. 10-17). On the x-axis the NO/NO_2 ratios are plotted, and on the y-axis the corresponding NO values are plotted.

The regression lines are drawn through the points; these lines are $y = m_1x + b_1$ and $x = m_2y + b_2$, with

$$m_1 = \frac{\Sigma xy - \frac{\Sigma x . \Sigma y}{n}}{\Sigma x^2 - \frac{(\Sigma x)^2}{n}} \quad , \quad m_2 = \frac{\Sigma xy - \frac{\Sigma x . \Sigma y}{n}}{\Sigma y^2 - \frac{(\Sigma y)^2}{n}}$$

$$b_1 = \frac{\Sigma y}{n} - m_1 \frac{\Sigma x}{n} \quad , \quad b_2 = \frac{\Sigma x}{n} - m_2 \frac{\Sigma y}{n}$$

The correlation coefficient $R = \dfrac{\sum xy - \dfrac{\sum x \sum y}{n}}{\sqrt{\left\{\sum x^2 - \dfrac{(\sum x)^2}{n}\right\}\left\{\sum y^2 - \dfrac{(\sum y)^2}{n}\right\}}}$

These functions are calculated per quarter and per combination of quarters. The graph shows a better match for the winter quarters than the summer quarters. The correlation is highest for the entire sampling period.

The findings of the temperature chart can be observed here again; that is, in the winter quarters the NO levels and the ratios are higher than in the summer quarters (compare the intersections of the regression lines).

7. Provisional Conclusions

This report has to be limited to provisional conclusions because the mobile units' data are only a small part of the data obtained in the project South Holland. Only the data of NO and NO_2 are (manually) processed.

7.1. General

1. In the measuring program used, which is very labor intensive, not enough use is made of the possibilities offered by automated instruments. Each working day, between 8 a.m. and 5 p.m., six sampling series are obtained; much information is lost this way.

2. The above motivates the following suggestions:

   a. Visit locations for a longer period.

   b. Sample for 24 hours a day.

   c. Sample multiple components, for example, hydrogen carbons, dust (which contains for example lead) and the meteo factors such as temperature, wind direction and velocity, relative humidity and possibly radiation.

   d. Replace the Euroglas-ozone monitor.

3. The program was quite satisfactory for the period under consideration as a random system. Notwithstanding some degrees of freedom, the distribution of the samples is quite good with respect to the days of the week, time of the day, and even temperature and wind direction.

4. A sampling program with mobile units in addition to stationary sampling stations is certainly appropriate, especially when information about a certain region has to be obtained in a rather short period.

7.2. Results of the Measurements

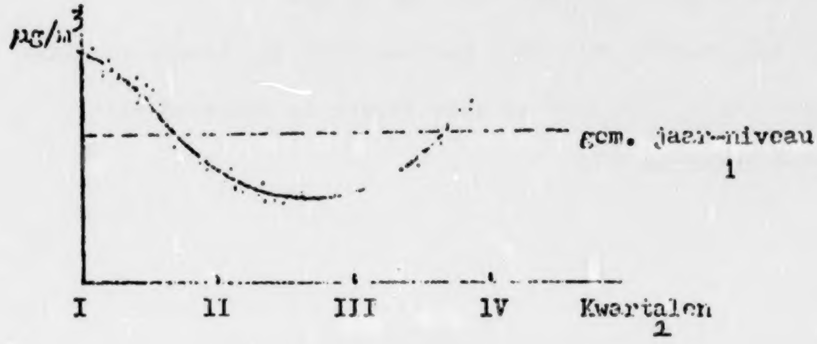
1. The iso-lines show highest NO and NO_2 levels in the city centers. The highest NO levels were observed at sampling locations near highways. This is not observed in Schiedam because no sampling points were located in the city center.

2. The NO levels in the winter half year are considerably higher than in the summer half year. To a lesser degree, this applies also to NO_2.

3. The NO/NO_2 ratio has a significant correlation with the NO level.

4. The NO level, and therefore also the NO/NO_2 ratio, is temperature dependent. Although the NO_2 fluctuations are smaller than those of NO, there are indications that a kind of coupled reaction $NO \rightarrow NO_2$ exists. The divergences of the NO and NO_2 levels differ a factor 2.

5. The movements of the NO_x levels in a year can be represented by a macro wave for the average quarter levels, with a superposition of a micro wave for the daily variations and with a "valley" at the middle of the day due to temperature dependence.



key: 1. average year level

2. quarters

6. An attempt is made to test the following rule of thumb: relative contributions of Industry:Traffic:Home Heating = 1/3:1/3:1/3. The sector "I", which contains the entire industry region, has a relative contribution of 24% to the NO level and 26% to the NO_2 level. No pre-load was considered. More pre-load will cause the relative contributions to increase.

7. The same attempt (see 6. above) is made for Vlaardingen. Here the relative "I" contribution for NO was 54% and for NO_2 52%. However, it is striking that the absolute levels of both NO and NO_2 in Vlaardingen are lower than those of the entire region.

8. The information that can be obtained from the NO/NO_2 ratio is still unclear. The great dependence on the NO level makes it hard to find, for example the distance from the sampling location to the source based on the reaction time that is necessary to form NO_2. NO_2 lags so much behind NO that one wonders: Where does the NO go?

9. Other measurements show that the measured NO_2 levels in this report are rather high, 52 $\mu g/m^3$ in this report as compared with 30-40 $\mu g/m^3$ from other sources.

/Translator's note: Please note the European convention of denoting the decimal point with a comma in the following Figures and Tables./

Tables

Table 1: Data of the Measurements Network from the Mobile Unit of KvW/DCMR

Key:
1. Internal Serial Code
2. R.I.V. Code
3. Center of gravity of region

Table 2: Frequency of Measurements of NO and/or NO_2 on Weekdays for the Three Routes

Key:
1. Total/R
2. Total/time
3. Monday
4. Tuesday
5. Wednesday
6. Thursday
7. Friday

Table 3: NO Averages in $\mu g/m^3$, Computed per Period for Each measuring Point and for the Entire Region

Key:
1. Measuring location
2. Entire region

Table 4: NO_2 Averages in $\mu g/m^3$, Computed per Period for Each Measuring Point and for the Entire Region

Key: See Table 3 for key.

Table 5: Averages for the Ratio of (Paired) NO/NO_2, Computed per Period for Each Measuring Point and for the Entire Region

Key: See Table 3 for key.

Table 6: Averages, Deviations and Relative Contribution Percentages of all Samplings Together for the Sectors "I", "K" and "R"

Note: Eight series of samples are omitted because of the partial overlapping "I" and "K" at Hoogvliet. See also Figs. 20 and 21.

Key: 1. Quarter
2. Year

Table 7: Contributions in Percentages to the NO and NO_2 Levels of "I", "K" and "R" at Three Locations in Vlaardingen; the Same for These Three Locations Together. (1973; pre-load equals zero.) Next to the averages the intervals are given for $p=0.05$

Table 8: Daily NO Levels

Key: 1. (Average of the entire region)
2. Hour
n: number of samples
m: average
sd: standard deviation
kwart: quarter

Table 9: Daily NO_2 Levels

Use key for Table 8.

Table 10: Daily NO/NO_2 Levels

Use key for Table 8.

Table 11: Temperature/NO Level

Key:
1. Average for the entire region
2. Temperature class

n, m and sd: See Table 8.

Table 12: Temperature/NO_2 Level

Key:
1. Average for 18 sampling locations

n, m and sd: see Table 8.

Table 13: Temperature/Level of NO/NO_2

Use key for Table 12.

Table 14: Averages and Deviations for the Entire Region per Quarter and Combinations of Quarters

Key:
1. Measuring location
2. Entire region (18 locations)
3. Paired
4. Number
5. Average
6. Standard deviation
7. Period
8. Quarter

Tabel : 1

Gegevens van het meetnet van de meetauto van KvW/DCMR

Intern volgnr. 1	R.I.V.-kode 2	Coördinaten	Adres	Sektor "K"	Sektor "I"	Route
14	19 13 9	43810850	Sweelincksingel,Schiedam-Groenoord	280-320	170-280	3
15	19 14 7	43800869	Klaas Katerstraat,Schiedam	280-300	190-280	1
17	19 14 8	43800910	van Goensstraat,Rotterdam-Blijdorp	280-300	220-280	1
18	20 14 9	43600930	Lombardkade,Rotterdam-Centrum	270-290	230-270	2
21	20 13 2	43590810	Geert Grootelaan,Vlaardingen-Zuidbuurt	300-340	110-290	1
23	20 13 3	43600850	Schiedamsedijk,Vlaardingen-Oost	290-320	160-280	2
25	20 14 1	43620890	Benj.Franklinstraat,Rotterdam-West	280-300	220-280	2
26	20 14 2	43600909	Rochusserstraat,Rotterdam	280-300	230-280	3
27	20 14 3	43610932	Meeuwenstraat,R'dam-Noordereiland	280-300	240-280	2
28	20 15 1	43630947	Feijenoorddijk,R'dam-Feijenoord	270-290	240-270	1
32	20 14 4	43390867	Pastoriedijk,Rotterdam-Pernis	290-320	230-290	1
35	20 14 6	43400930	Voetjesstraat,Rotterdam-Zuid	280-300	260-280	1
36	20 15 4	43400950	Immobilialaan,R'dam-Hillesluis	290-300	260-290	3
38	20 13 6	43180832	Ferd.Huyckstraat,Rotterdam-Hoogvliet	310-340	270- 70	*) 2
39	20 14 7	43200852	Hoefsmidstraat,Rotterdam-Hoogvliet	300-330	270- 40	*) 3
42	20 14 5	43200909	Schulpweg,Rotterdam-Charlois	290-310	260-290	2
43	20 14 9	43200930	Slinge, Rotterdam-Zuidwijk	290-310	270-290	3
50	19 13 8	43650322	Floreslaan,Vlaardingen-Ind.Buurt	290-330	120-260	3
Z	-	43530688	Zwaartepunt Meetgebied 3	(290-320)	(230-280)	-

*) Hier vallen "K" en "I" samen !

Tabel 2

Meetfrekwentie op weekdagen, waarop NO en/of NO_2 is gemeten, onderverdeeld naar de drie routes.

		IV-'72			I-'73			II-'73			III-'73			IV-'73			1973			
	R	R1	R2	R3	R1	R2	R3	R1	R2	R3	R1	R2	R3	R1	R2	R3	R1	R2	R3	Tot
MA 3	1	2			12			12			12			6			42			140
	2		0			6			12			17			6			41		
	3			0			18			12			15			12			57	
DI 4	1	18			12			6			16			17			51			157
	2		4			24			6			5			18			53		
	3			0			6			12			17			18			53	
WO 5	1	0			18			6			12			16			52			153
	2		11			12			18			6			12			48		
	3			16			12			18			11			12			53	
DO 6	1	18			6			21			16			17			60			168
	2		14			24			18			0			6			48		
	3			12			18			18			18			6			60	
VR 7	1	0			13			16			10			8			52			138
	2		6			6			15			24			12			57		
	3			11			11			12			0			6			29	
Tot/R 1		38	35	39	66	72	65	61	69	72	66	52	61	64	54	54	257	247	252	
Tot/tijdv. 2		112			203			202			179			172			756			

Tabel 3

Gemiddelden voor NO, berekend per periode voor elk meetpunt en het gehele meetgebied. (NO in $\mu g/m^3$)

meetpunt nr.:	4e Kw 1972:	1e Kw 1973:	2e Kw 1973:	3e Kw 1973:	4e Kw 1973:	IV-'72 +I-'73	II+III 1973 :	1973
14	96,2	111,4	26,3	30,0	62,2	106,0	27,9	56,4
15	104,0	63,6	21,4	19,3	44,6	77,9	20,4	37,5
17	120,5	68,4	24,9	25,1	55,7	86,8	25,0	43,7
18	52,6	75,8	26,8	32,8	90,8	67,3	29,4	64,6
21	89,7	52,7	29,4	33,0	55,6	72,3	30,4	41,2
23	96,8	74,3	25,8	21,7	72,2	83,3	24,0	49,3
25	69,8	66,6	28,2	26,6	89,7	67,7	27,5	53,2
26	107,6	155,8	37,1	52,6	103,3	137,1	44,9	86,5
27	92,5	84,3	24,3	27,7	91,7	87,1	25,8	56,5
28	104,8	70,0	24,4	17,8	49,4	82,3	21,1	40,9
32	64,3	51,2	30,1	17,6	47,8	55,8	23,6	36,5
35	70,0	81,3	26,0	15,9	79,3	77,8	20,7	51,2
36	69,0	109,0	24,5	17,9	88,6	94,0	21,5	58,6
38	69,7	69,6	21,0	35,6	55,2	69,6	27,1	46,2
39	89,3	113,6	18,5	23,6	47,8	103,6	20,7	50,0
42	88,3	80,9	26,5	31,6	86,1	83,9	28,6	57,3
43	90,3	125,7	39,7	29,4	75,9	111,1	34,7	65,2
50	63,5	82,5	22,3	19,4	64,8	75,4	21,0	45,5
gehele meetgebied:	85,6	84,4	26,6	26,8	68,2	84,8	26,7	51,8

2

Tabel 4

Gemiddelden voor NO_2 in µg/m^3, berekend per periode voor elk meetpunt en het gehele meetgebied.

meetpunt nr:	4e Kw 1972:	1e Kw 1973:	2e Kw 1973:	3e Kw 1973:	4e Kw 1973:	IV-'72 +I-'73	II+III 1973:	1973
14	65,7	59,5	36,6	55,6	53,4	61,8	44,4	50,0
15	59,3	46,4	42,3	50,4	53,9	50,9	46,3	48,0
17	49,0	48,9	43,4	70,4	58,4	48,9	58,9	56,2
18	80,1	52,6	49,7	82,2	78,9	63,6	63,6	63,7
21	57,0	42,0	26,5	40,7	62,5	47,3	33,6	43,0
23	69,2	48,8	42,1	62,4	73,0	57,5	51,7	55,6
25	70,5	56,7	46,7	62,1	79,1	62,2	52,9	59,7
26	78,0	65,8	49,5	88,2	66,0	70,8	67,9	66,9
27	68,7	57,9	45,8	61,0	71,2	61,5	52,3	58,0
28	61,7	53,9	39,8	52,1	49,8	56,9	45,7	49,0
32	74,2	41,3	29,5	38,4	42,1	52,9	33,9	37,8
35	66,3	52,4	39,3	48,9	55,1	57,8	44,3	49,1
36,	62,8	63,0	41,0	54,4	53,0	62,9	47,1	52,3
38	54,9	38,8	34,6	59,3	66,1	45,3	45,0	47,9
39	48,3	46,1	31,6	54,6	47,2	46,9	41,5	44,0
42	64,0	48,8	38,1	58,0	70,6	54,4	46,5	52,6
43	69,9	59,4	42,2	63,4	52,1	63,7	52,3	54,0
50	57,0	54,8	28,8	73,3	55,9	55,6	46,6	51,0
gehele meetgebied: 2	63,6	51,8	39,3	59,4	60,1	56,3	48,5	53,1

Tabel 5

Gemiddelden voor de verhouding van(gepaard) NO/NO_2,

berekend per periode voor elk meetpunt en het gehele meetgebied.

$\overline{(NO/NO_2)}$

meetpunt nr:	4e Kw 1972:	1e Kw 1973:	2e Kw 1973:	3e Kw 1973:	4e Kw 1973:	IV+'72 +I-'73	II+III 1973:	1973:
14	1,54	1,94	0,89	0,87	1,09	1,79	0,88	1,18
15	1,67	1,29	0,60	0,37	0,90	1,42	0,48	0,80
17	2,91	1,34	0,55	0,40	1,02	1,90	0,47	0,83
18	0,70	1,41	0,53	0,46	1,21	1,15	0,50	0,92
21	1,51	1,37	0,97	0,58	1,07	1,46	0,78	1,00
23	1,38	1,54	0,59	0,44	1,07	1,47	0,52	0,95
25	0,96	1,19	0,57	0,46	0,98	1,12	0,52	0,83
26	1,62	2,25	0,75	0,55	1,43	1,99	0,66	1,25
27	1,09	1,43	0,56	0,57	1,75	1,31	0,56	1,07
28	1,65	1,29	0,65	0,34	1,20	1,42	0,50	0,89
32	0,92	1,19	1,03	0,38	1,13	1,09	0,71	0,94
35	0,99	1,44	0,67	0,33	1,66	1,30	0,50	1,05
36	1,10	1,57	0,61	0,38	1,46	1,38	0,50	0,96
38	1,33	1,85	0,65	0,51	0,87	1,64	0,59	1,03
39	1,70	2,60	0,68	0,48	1,02	2,31	0,59	1,19
42	1,37	1,58	0,67	0,57	1,17	1,50	0,63	1,04
43	1,36	2,02	0,98	0,95	1,49	1,75	0,96	1,33
50	1,50	1,53	0,99	0,32	1,12	1,52	0,72	1,02
Gehele meetgebied:	1,41	1,59	0,73	0,50	1,21	1,52	0,62	1,02

		NO				NO_2			
		Tot.	"I"	"K"	"R"	Tot.	"I"	"K"	"R"
1^e Kw.'73 1	$\overline{x}$	84,4	70,2	79,4	93,4	51,8	53,8	47,9	51,7
	r	± 8,0	±10,6	±19,2	±12,1	± 2,4	± 4,8	± 5,6	± 3,1
	%	100	25,5	12,8	61,7	100	32,5	12,6	54,9
2^e Kw.'73	$\overline{x}$	26,7	26,8	22,5	27,2	39,3	41,8	27,6	40,2
	r	± 2,3	± 5,0	± 9,2	± 2,9	± 2,2	± 3,9	± 4,8	± 2,7
	%	100	21,3	8,1	70,6	100	22,0	6,7	71,3
3^e Kw.'73	$\overline{x}$	26,8	25,3	18,9	28,2	59,4	48,7	60,6	61,9
	r	± 4,0	±15,3	± 9,2	± 3,8	± 5,6	± 9,4	±21,4	± 6,9
	%	100	17,0	6,8	76,2	100	14,6	10,0	75,4
4^e Kw.'73	$\overline{x}$	69,7	61,4	38,4	83,2	60,0	65,2	49,0	60,3
	r	±10,5	±14,2	± 8,3	±16,9	± 4,8	±11,3	± 7,7	± 5,6
	%	100	25,8	8,7	65,5	100	33,0	12,6	54,4
Jaar 1973 2	$\overline{x}$	51,8	50,1	43,7	54,0	52,1	53,4	46,3	52,7
	r	± 2,2	± 6,5	± 8,4	± 5,3	± 2,0	± 4,1	± 5,5	± 2,5
	%	100	23,9	10,2	65,9	100	25,8	10,7	63,5

Tabel 6. Gemiddelden en spreidingen (p=0,05), alsmede de relatieve bijdragepercentages voor alle meetpunten tezamen voor de sektoren "I", "K" en "R".

Van het cijfermateriaal zijn in totaal 8 waarnemingsseries niet opgenomen, omdat op de twee meetpunten in Hoogvliet "I" en "K" gedeeltelijk samenvallen.

Zie ook fig.20 en fig.21.

Tabel 7

De bijdragen in procenten van "I", "K" en "R" *) aan de niveaus van NO en NO_2 op drie meetplaatsen en de combinatie van deze drie meetplaatsen in Vlaardingen. (1973 ; voorbelasting is nul) Achter de gemiddelden zijn de intervallen vermeld, voor p= 0,05

		NO	NO_2
Floreslaan:	"I"	55,5 (33,9-74,3)	46,2 (35,3-59,2)
$\overline{NO}$ =45,6 µg/m^3	"K"	15,4 (7,3-30,1)	11,4 (6,7-18,1)
$\overline{NO_2}$=50,8 µg/m^3	"R"	29,1 (15,2-49,4)	42,4 (29,9-54,5)
		+	+
		100 %	100 %
Schiedamsedijk:	"I"	42,5 (23,6-65,4)	48,3 (27,2-63,7)
$\overline{NO}$ =49,3 µg/m^3	"K"	10,9 (4,0-24,0)	8,2 (4,8-14,9)
$\overline{NO_2}$=55,6 µg/m^3	"R"	46,6 (25,6-68,3)	43,5 (29,8-63,6)
		+	+
		100 %	100 %
Geert Grootelaan:	"I"	65,2 (37,2-88,6)	63,7 (43,8-78,8)
$\overline{NO}$ =42,0 µg/m^3	"K"	7,9 (1,7-23,2)	8,1 (4,3-15,7)
$\overline{NO_2}$=43,0 µg/m^3	"R"	26,9 (8,4-54,7)	28,2 (15,5-46,7)
		+	+
		100 %	100 %
VLAARDINGEN :	"I"	53,8 (40,1-67,2)	51,9 (42,4-60,7)
$\overline{NO}$ =45,6 µg/m^3	"K"	10,8 (6,4-17,5)	8,9 (6,4-12,4)
$\overline{NO_2}$=49,6 µg/m^3	"R"	35,4 (23,8-48,9)	39,2 (31,2-48,4)
		+	+
		100 %	100 %

*) "I", "K" en "R" zijn de afkortingen voor de windrichtingssektoren waarin resp. de Industrie, het kassengebied en de rest liggen, en die nader zijn aangegeven in tabel 1

Tabel 8

DAGVERLOOP NO niveau

(gemiddelde over het gehele meetgebied)

periode uur		4^e kwart. 1972	1^e kwart. 1973	2^e kwart. 1973	3^e kwart. 1973	4^e kwart. 1973
9	n	3	11	5	11	18
	m	36,67	104,82	42,40	44,00	105,28
	sd	14,57	96,16	6,62	25,10	100,48
10	n	13	20	26	22	28
	m	85,15	110,05	27,73	36,50	91,39
	sd	73,00	79,72	14,48	24,82	90,74
11	n	14	28	34	37	29
	m	119,14	106,93	29,26	31,73	76,10
	sd	91,22	62,05	20,48	43,21	70,44
12	n	20	30	31	29	24
	m	83,10	78,07	24,65	26,21	62,29
	sd	56,97	37,30	13,16	25,67	57,89
13	n	15	36	25	25	28
	m	90,00	69,83	24,40	20,56	52,54
	sd	75,06	47,50	19,86	15,53	37,44
14	n	16	33	32	25	30
	m	84,50	75,73	25,62	16,04	44,37
	sd	62,28	52,81	15,43	7,04	33,04
15	n	18	25	30	19	9
	m	66,11	77,48	24,67	20,47	63,56
	sd	5,3155,81	48,59	16,01	21,04	56,33
16	n	14	15	17	9	1
	m	101,07	72,47	26,47	14,67	84
	sd	99,70	45,56	16,17	8,83	-
17	n	4	2	1	2	-
	m	51,50	55,50	26	25,00	--
	sd	14,01	36,06	-	18,38	-

n: aantal waarnemingen, m: gemiddelde, sd: standaardafwijking tabel 9 DAGVERLOOP NO_2 niveau

(gemiddelde over het gehele meetgebied.)

n : aantalwaarnemingen, m: gemiddelde , sd: standaardafw.

periode uur 2		4 kwartaal 4e kwart. 1972	1 kwartaal 1e kwart. 1973	2 kwartaal 2e kwart. 1973	3 kwartaal 3e kwart. 1973	4e kawrt. 1973
	n	4	11	5	11	19
9	m	59,75	53,18	50,40	64,05	64,89
	sd	12,26	5,21	8,82	23,18	29,66
1i	n	14	21	26	21	28
10	m	58,21	62,95	40,65	57,19	60,81
	sd	23,38	24,24	15,60	29,89	28,52
	n	16	26	34	35	29
11	m	66,88	61,54	40,53	61,49	68,90
	sd	14,24	23,13	15,56	39,54	47,23
	n	21	31	30	29	25
12	m	64,05	47,13	38,73	61,93	61,68
	sd	19,61	10,39	11,52	33,46	47,23
	n	15	35	26	23	29
13	m	57,60	43,71	38,19	70,04	55,17
	sd	22,92	12,99	18,06	61,19	25,19
	n	17	33	32	24	30
14	m	69,12	48,46	40,84	55,50	55,30
	sd	30,50	15,42	19,67	27,19	26,36
	n	18	26	30	17	10
15	m	62,50	53,04	37,00	55,47	46,50
	sd	15,85	15,88	13,54	33,76	20,93
	n	14	15	17	9	1
16	m	74,79	51,33	35,24	33,89	72
	sd	29,09	12,36	16,06	19,20	-
	n	4	2	1	2	-
17	m	69,75	55,50	27	84,5	-
	sd	17,25	6,36	-	14,50	-

tabel 10 DAGVERLOOP NO/NO$_2$-verhouding
(gemiddeld over het gehele meetgebied)

periode uur 2		4^e kwart. 1972	1^e kwart. 1973	2^e kwart. 1973	3^e kwart. 1973	4^e kwart. 1973
9	n	3	11	5	11	18
	m	0,64	1,93	0,88	0,71	1,58
	sd	0,32	1,68	0,30	0,39	1,37
10	n	12	20	26	21	28
	m	2,07	1,71	0,69	0,62	1,63
	sd	2,11	0,76	0,29	0,24	1,70
11	n	14	27	34	35	28
	m	1,79	1,69	0,71	0,54	1,34
	sd	1,50	0,73	0,33	0,48	1,49
12	n	20	31	30	29	24
	m	1,32	1,65	0,64	0,42	1,01
	sd	0,87	0,70	0,24	0,25	0,76
13	n	15	34	25	23	28
	m	1,44	1,54	0,77	0,34	0,95
	sd	1,09	0,80	0,79	0,22	0,71
14	n	16	32	32	24	30
	m	1,35	1,48	0,71	0,37	0,87
	sd	1,04	0,80	0,45	0,29	0,81
15	n	18	25	30	17	9
	m	1,10	1,47	0,74	0,53	1,27
	sd	0,84	0,81	0,51	0,71	0,92
16	n	14	15	17	9	1
	m	1,44	1,53	0,85	0,82	1,17
	sd	1,22	1,28	0,61	1,17	-
17	n	4	2	1	2	-
	m	0,80	0,97	0,96	0,29	-
	sd	0,35	0,54	-	0,17	-

n: aantal waarnemingen m: gemiddelde
sd: standaardafwijking

Tabel 11

TEMPERATUURVERLOOP NO niveau

temp.klasse °C		4e kwart. 1972	1e kwart. 1973	2e kwart. 1973	3e kwart. 1973	4e kwart. 1973
	n	6	15			16
1	m	172,00	136,27			93,31
	sd	57,94	90,04			57,06
	n	4	59	12		13
4	m	128,75	98,84	43,75		104,92
	sd	81,07	53,66	25,85		95,88
	n	18	77	23		34
7	m	132,50	76,22	30,52		87,79
	sd	114,49	49,56	11,72		91,53
	n	48	43	35	1	41
10	m	77,71	65,58	34,77	54	69,90
	sd	53,16	50,12	17,07	-	62,97
	n	32	6	35	18	42
13	m	59,81	56,50	26,91	26,00	56,74
	sd	45,57	49,17	16,50	14,00	46,76
	n	8		37	67	21
16	m	50,00		24,83	23,13	24,95
	sd	23,05		17,46	18,74	10,42
	n			30	50	
19	m			18,40	29,56	
	sd			7,01	42,08	
	n			21	24	
22	m			17,14	24,46	
	sd			10,31	18,83	
	n			7	12	
25	m			15,71	32,50	
	sd			4,07	26,88	
	n			1	6	
28	m			9	22,83	
	sd			-	10,07	

gemiddelde over het gehele meetgebied

n: aantal waarnemingen

m: gemiddelde

sd: standaardafwijking

TABEL 12 TEMPERATUURVERLOOP NO_2 NIVEAU

temp.klasse °C		4e kwart. 1972	1e kwart. 1973	2e kwart. 1973	3e kwart. 1973	4e kwart. 1973
	n	6	12			16
1	m	72,67	49,00			75,94
	sd	12,19	15,80			41,58
	n	4	61	12		13
4	m	46,75	53,03	43,00		100,46
	sd	4,57	15,39	14,08		37,94
	n	18	78	24		37
7	m	60,89	49,73	36,63		60,89
	sd	18,31	13,98	14,95		38,35
	n	64	43	35	1	42
10	m	63,02	52,93	44,37	72	50,38
	sd	24,10	13,98	13,67	-	14,82
	n	34	6	35	17	42
13	m	65,85	64,17	26,91	54,00	50,00
	sd	20,02	35,96	16,50	15,91	19,47
	m	8		37	67	21
16	m	82,75		38,62	50,44	62,10
	sd	26,00		15,26	23,53	25,00
	n			30	49	
19	m			39,53	52,15	
	sd			13,88	34,55	
	n			21	22	
22	m			31,57	85,82	
	sd			14,15	61,61	
	n			7	9	
25	m			30,00	76,27	
	sd			16,00	44,52	
	n				6	
28	m				99,58	
	sd				46,33	

gemiddelde over 18 meetpunten,
n : aantal waarnemingen
m : gemiddelde
sd : standaardafwijking

tabel 13 TEMPERATUURVERLOOP NO/NO_2 VERHOUDING

temp.klasse °C		4^e kwart. 1972	1^e kwart. 1973	2^e kwart. 1973	3^e kwart. 1973	4^e kwart. 1973
1	n	6	12			16
	m	2,41	2,74			1,29
	sd	0,83	1,80			0,53
4	n	4	59	12		13
	m	2,67	1,88	1,07		1,15
	sd	1,55	0,70	0,56		1,44
7	n	18	77	23		34
	m	2,17	1,47	0,98		1,68
	sd	1,62	0,72	0,55		1,92
10	n	47	43	35	1	40
	m	1,39	1,18	0,82	0,75	1,30
	sd	1,22	0,60	0,38	-	0,98
13	n	32	6	35	17	42
	m	0,90	0,80	0,72	0,46	1,16
	sd	0,54	0,26	0,52	0,20	0,82
16	n	8		37	67	21
	m	0,63		0,70	0,47	0,42
	sd	0,29		0,56	0,26	0,14
19	n			30	48	
	m			0,49	0,57	
	sd			0,18	0,51	
22	n			21	22	
	m			0,56	0,44	
	sd			0,29	0,64	
25	n			7	9	
	m			0,59	0,73	
	sd			0,15	1,13	
28	n				6	
	m				0,24	
	sd				0,06	

gemiddelde over 18 meetpunten

n : aantal waarnemingen
m : gemiddelde
sd : standaardafwijking

Dienst Centraal Milieubeheer
Rijnmond
Stationsplein 2,Schiedam
010- 150 800

Proefproject Zuid-Holland , [illegible]

Primaire Verwerking Meetresultaten

Tabel 14

Meetplaats(en):[1] GEHELE MEETNET[2] (18 meetpunten)

Periode[7] :	NO			NO₂			$\overline{NO}$ / $\overline{NO_2}$	(gepaarde)[3] NO/NO₂		
	n[4]	gem[5]	s.d.[6]	n	gem	s.d.		n	gem	s.d.
4ᵉ kwartaal[8] 1972	117	85,56	71,24	123	63,58	20,07	1,35	117	1,41	1,21
1ᵉ kwartaal 1973	201	84,42	57,65	200	51,82	17,21	1,63	197	1,59	0,88
2ᵉ kwartaal 1973	201	26,55	16,66	201	39,25	15,73	0,68	200	0,73	0,47
3ᵉ kwartaal 1973	178	26,76	27,05	172	59,38	37,70	0,45	171	0,50	0,47
4ᵉ kwartaal 1973	167	69,54	68,17	171	60,08	31,36	1,16	166	1,21	1,19
4ᵉ kw '72 + 1ᵉ kw '73	318	84,84	62,88	323	56,29	19,19	1,51	314	1,52	1,02
4ᵉ kw '72 + 1ᵉ en 4ᵉ kw '73	485	79,57	65,09	494	57,61	24,15	1,38	480	1,42	1,09
2ᵉ en 3ᵉ kwartaal 1973	379	26,65	22,12	373	48,53	29,79	0,55	371	0,62	0,48
4ᵉ kw '72 + 1973	864	56,36	57,28	867	54,56	25,30	1,03	851	1,07	0,96
1973	747	51,78	53,40	744	53,07	25,78	0,98	734	1,02	0,91

Figures

Fig. 1: Map of Region with Sampling Locations and Routes

Fig. 2 Frequency of NO and/or NO_2 Measurements at Various Hour Intervals

Key:
1. Time interval
2. Weekdays
3. Measuring frequency
4. Wind frequency
5. Wind direction

Fig. 3 Frequency of NO and/or NO_2 Measurements at Various Temperature Intervals

See key for Fig. 2.

Fig. 4 Frequency of NO and/or NO_2 Measurements at Various Weekdays

See key for Fig. 2.

Fig. 5 Frequency of NO and/or NO_2 Measurements for Various Directions of Wind

Fig. 6 NO Iso-lines for 1973

Fig. 7 NO_2 Iso-lines for 1973

Fig. 8 Paired NO/NO_2 Iso-lines for 1973

Fig. 9	Levels of NO and NO_2 for the Entire Sampled Region per Quarter and per Season. The Same for Paired NO/NO_2 Ratios. Comment: the averages for 1973 are plotted as horizontal lines.
Fig. 20	Cumulative Relative Contribution Percentages to the Total NO Level of the Entire Sampling Region Key: 1. Note: the percentages are given for each quarter and for each season.
Fig. 21	Cumulative Relative Contribution Percentages to the Total NO_2 Level of the Entire Sampling Region Key: See key for Fig. 20.
Figs. 10-17	Regression Lines for NO and NO/NO_2 for Various Periods Key: kwartaal - quarter
Fig. 18	NO (——) and NO_2 (----) in 1973 for measuring point Sweelincksingel. The samples are averaged per 30 sector. The number of samples is indicated in each sector: NO/NO_2. The measured data during variable wind and the wind velocity are not considered.
Fig. 19	The same as Fig. 18 for the sampling location Schulpweg.

Figs. 20-21	See Fig. 9 (all three figures are on the same page.)
Fig. 22-31	Triangle Diagrams for the Contributions in Vlaardingen (Each Fig. is for a different location in Vlaardingen.) Fig. 26: On the axes the extreme values of NO for the three Vlaardingen locations are shown ($p=0.05$). Inside the large hexagon the derived hexagon of the combination is drawn. The dotted triangle shows the averages of the individual locations. Inside this triangle the average of the combination is located.
Figs. 32-39a	Daily NO and NO_2 Levels and NO/NO_2 Ratios Key: "geheel 1973": entire 1973
Fig. 40	3D Graph of NO/NO_2 Ratios, Time of the Day and Quarters of the Year Key: 1. Ratio 2. Quarter 3. Time

Figs. 41-48 Temperature/Level of NO and NO_2

Key: "geheel 1973" - the entire 1973

Figs. 49-57 Temperature Level of NO/NO_2 Ratio

Key: "geheel 1973" - the entire 1973

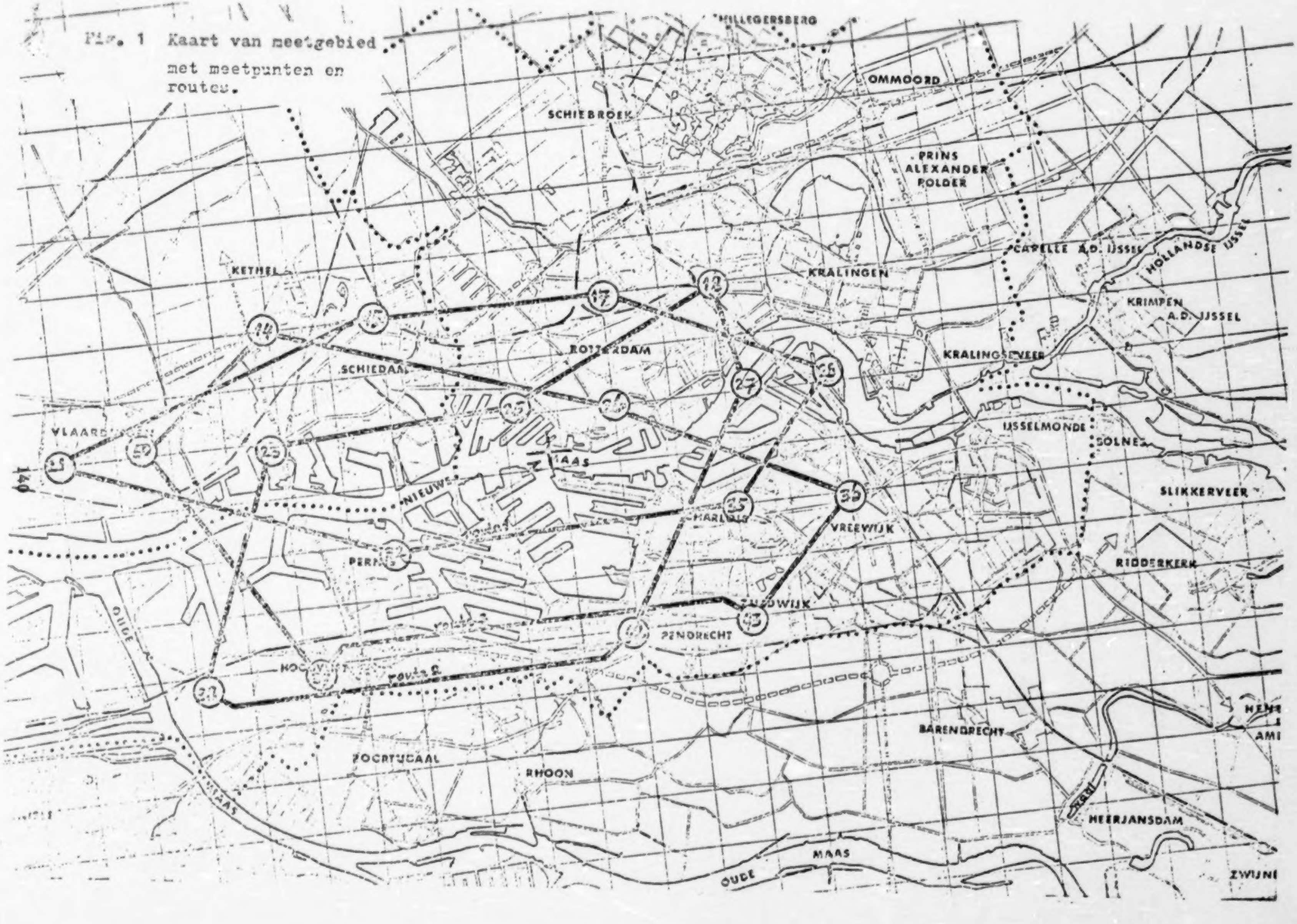


Fig. 1 Kaart van meetgebied met meetpunten en routes.

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KETHEL
KRALINGEN
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SOLNES
SLIKKERVEER
VLAARD
NIEUWE
MAAS
PERNIS
VREEWIJK
RIDDERKERK
ZUIDWIJK
PENDRECHT
OUDE
BARENDRECHT
ZOCRTUGAAL
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HEERJANSDAM
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Fig.2 aantal malen dat NO en/of NO_2 is gemeten in versch. uurintervallen.

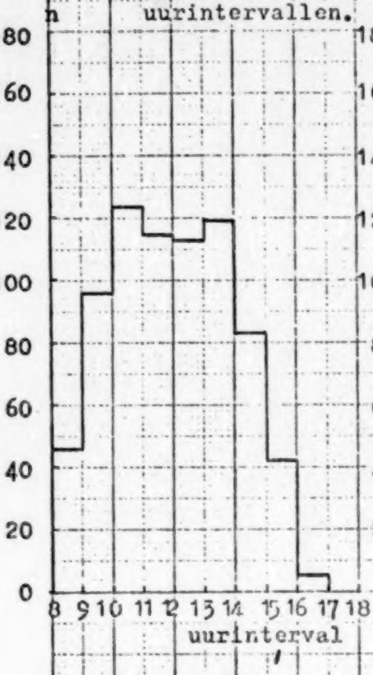


Fig.3 aantal malen dat NO en/of NO_2 is gemeten bij versch. temp.intervallen

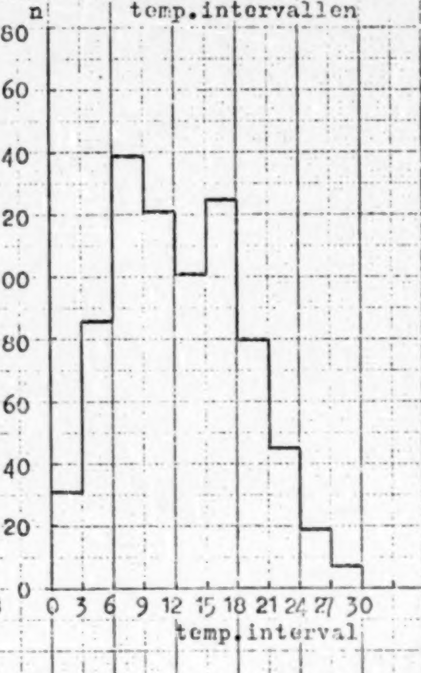


Fig.4 aantal malen dat NO en/of NO_2 is gemeten op de versch. weekdagen

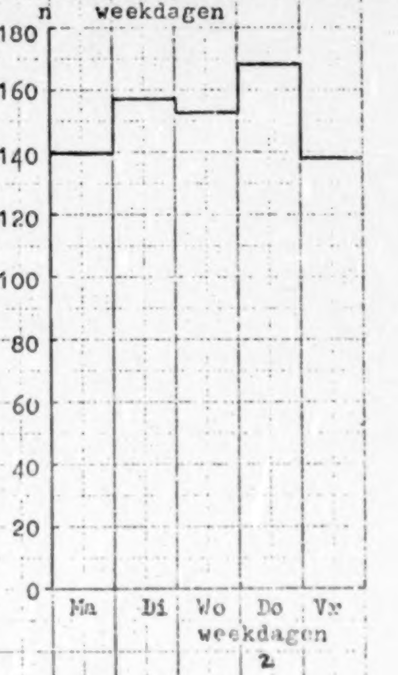


Fig. 5 Aantal malen dat NO en/of NO_2 is gemeten bij verschillende windrichtingen.

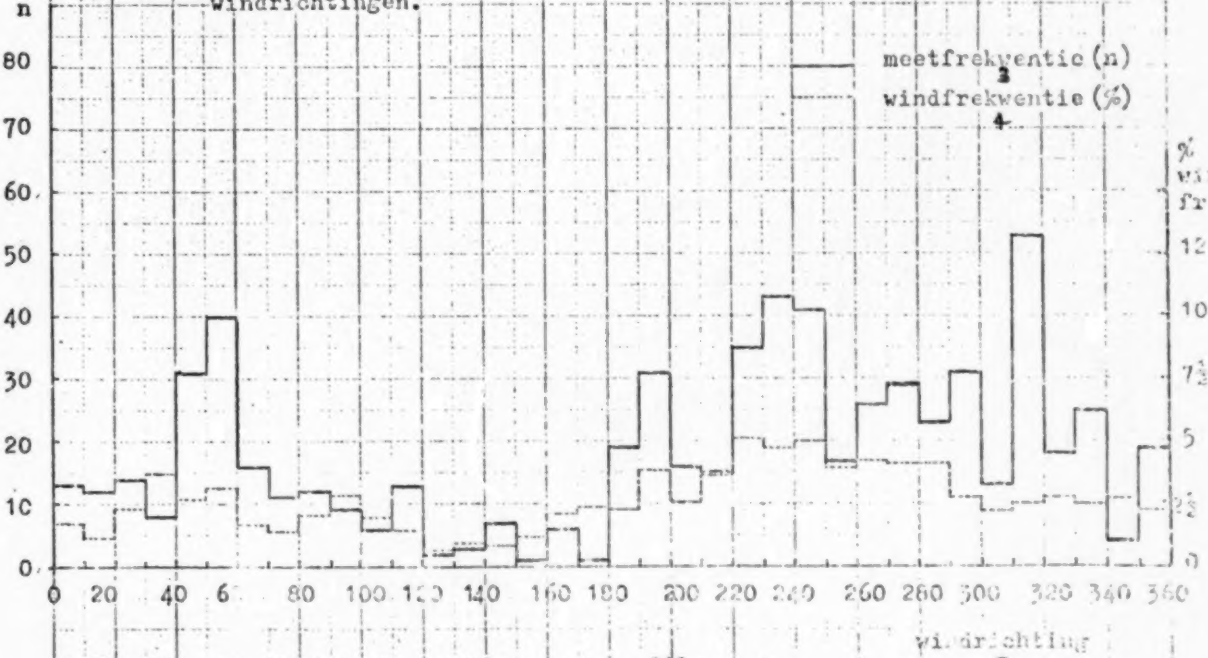


Fig. 6

NO-isolijnen voor 1973

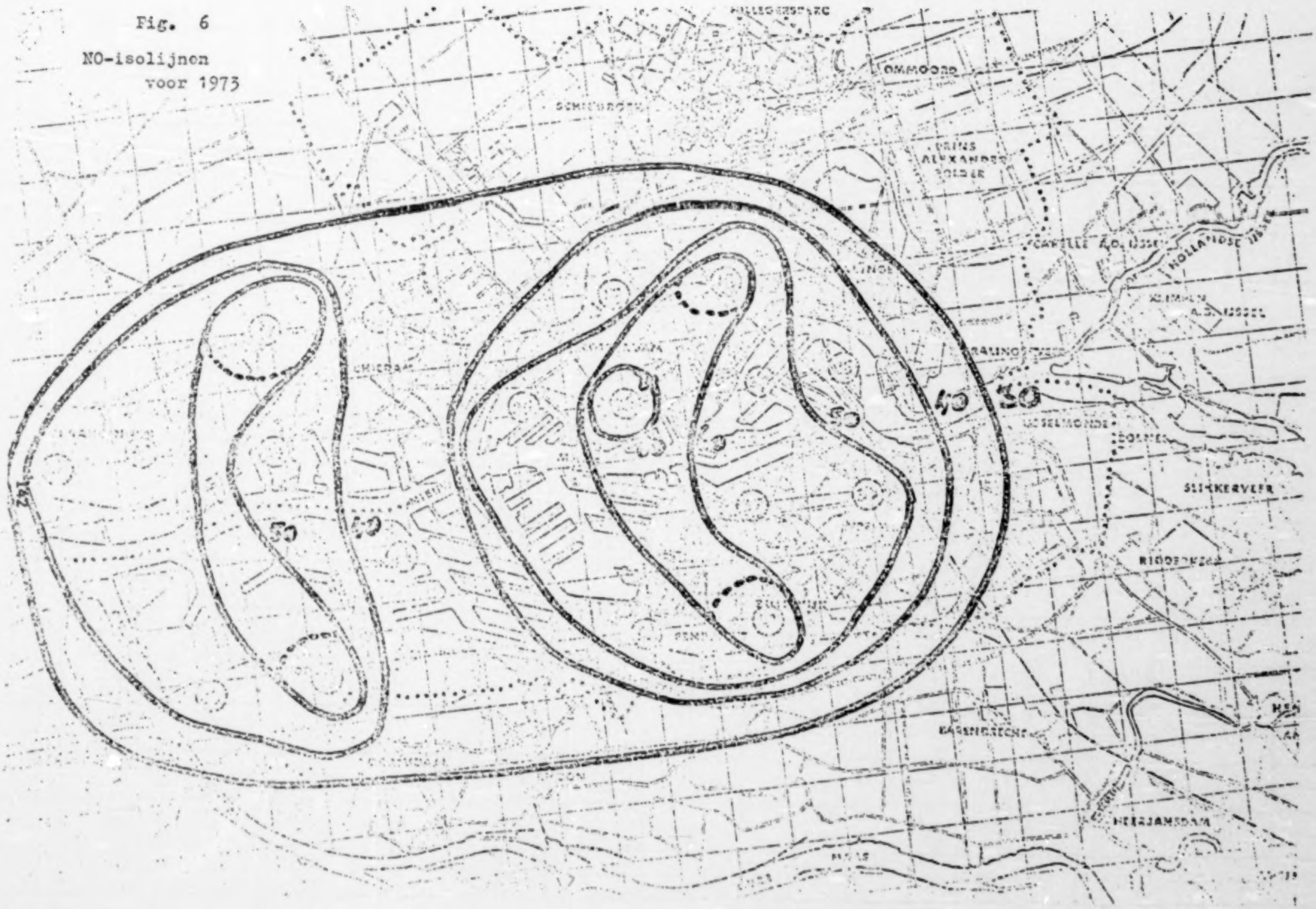

OMMOORD
SCHIEBROEK
PRINS ALEXANDERPOLDER
CAPELLE A.D. IJSSEL
HOLLANDSE IJSSEL
KRIMPEN A.D. IJSSEL
IJSSELMONDE
SLIKKERVEER
RIDDERKERK
BARENDRECHT
HEERJANSDAM
MAAS
30
40
50
60
50
60

Fig. 7

NO$_2$-isolijnen voor 1973

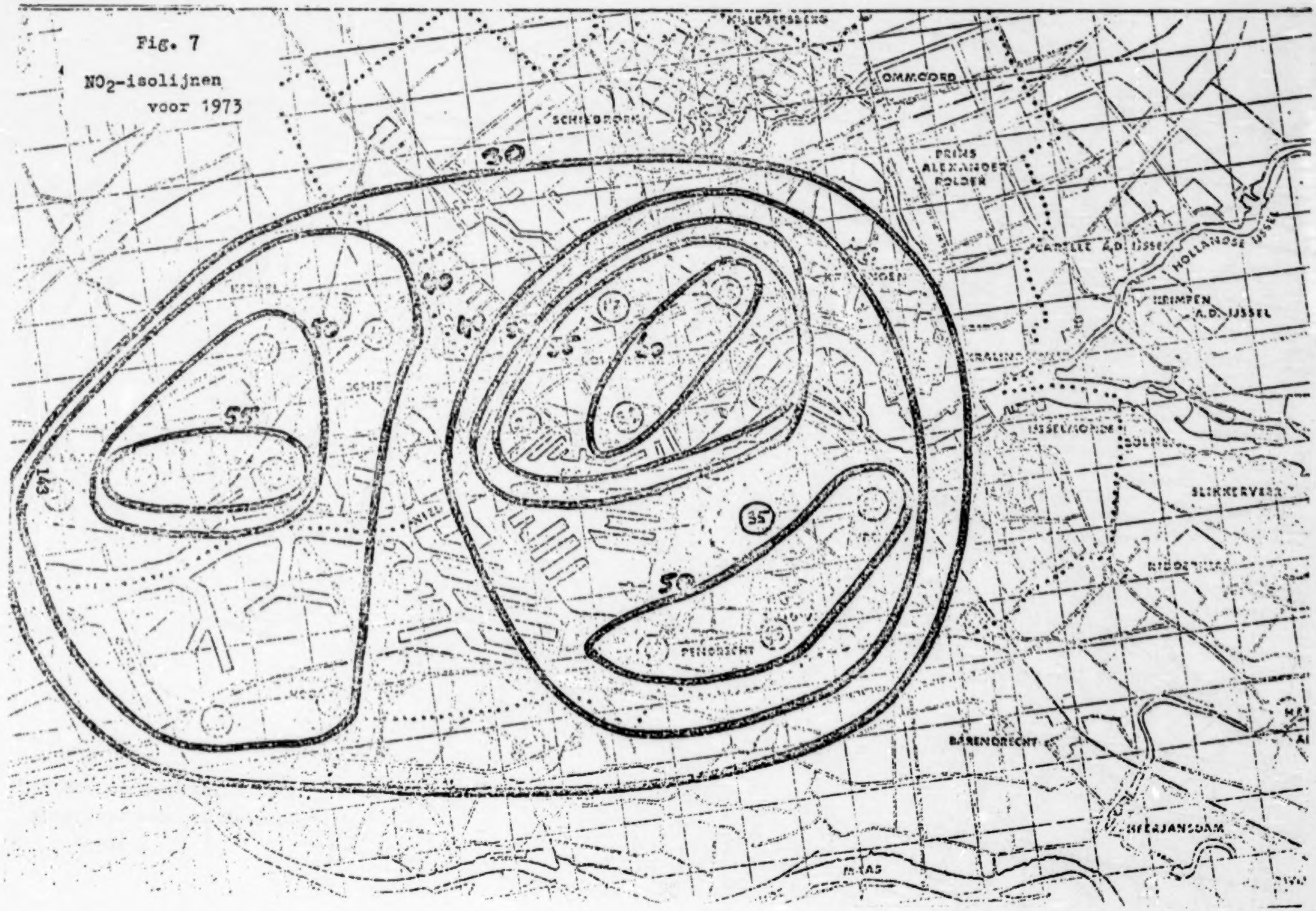
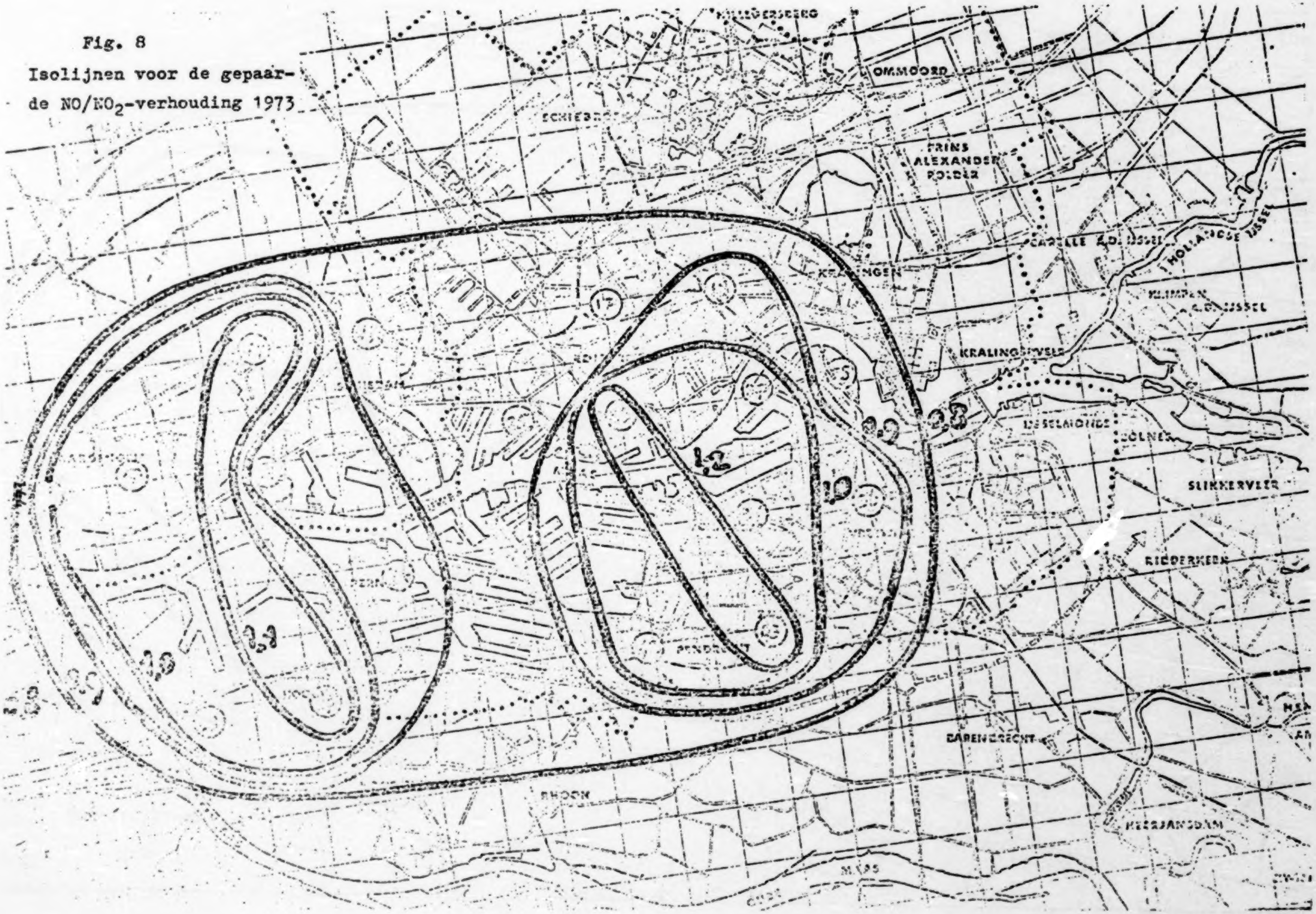

HILLEGERSBERG
OMMOORD
SCHIEBROEK
PRINS ALEXANDER POLDER
CAPELLE A.D. IJSSEL
HOLLANDSE IJSSEL
KRIMPEN A.D. IJSSEL
KRALINGEN
IJSSELMONDE
BOLNES
SLIKKERVEER
RIDDERKERK
BARENDRECHT
HEERJANSDAM
MAAS
PENDRECHT
30
50
55
50
55
60
35
50
17

Fig. 8

Isolijnen voor de gepaarde NO/NO_2-verhouding 1973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OMMOORD
SCHIEBROEK
PRINS ALEXANDER POLDER
CAPELLE A.D. IJSSEL
HOLLANDSE IJSSEL
KRALINGEN
KRIMPEN A.D. IJSSEL
KRALINGSEVEER
IJSSELMONDE
BOLNES
SLIKKERVEER
RIDDERKERK
PENDRECHT
BARENDRECHT
RHOON
HEERJANSDAM
MAAS
0,8
0,9
1,0
1,2
1,0
0,9
0,8

Fig. 9
Niveaus van NO en NO_2 van het gehele meetgebied per kwartaal en per seizoen.
Idem voor de verhouding van gepaarde NO/NO_2.
De gemiddelden voor 1973 staan als horizontale lijnen aangegeven.

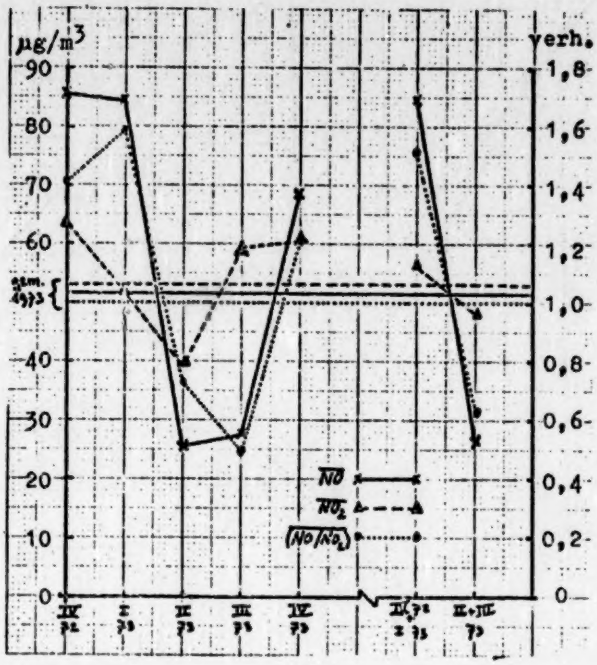


Fig. 20
Cumulatieve relatieve bijdragepercentages aan het totale niveau van NO in het gehele meetgebied.
Deze zijn aangegeven per kwartaal en per seizoen.

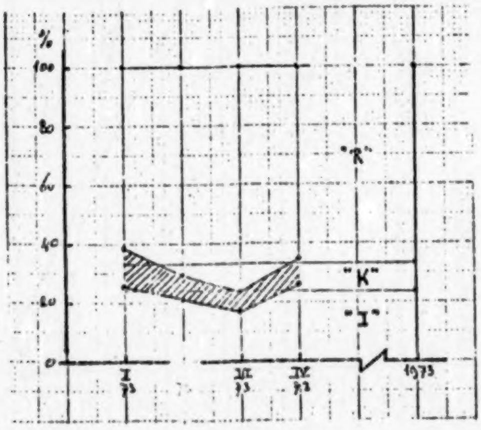


Fig. 21
Cumulatieve relatieve bijdragepercentages aan het totale NO_2-niveau in het gehele meetgebied
Deze zijn aangegeven per kwartaal en per seizoen.

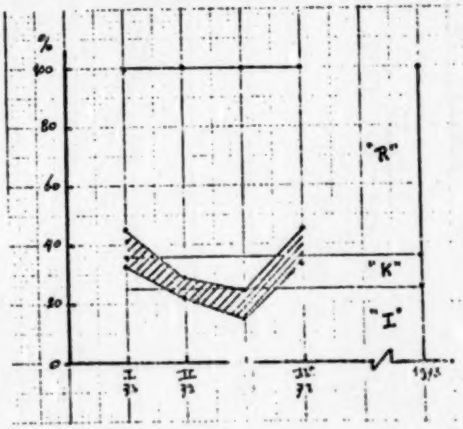


Fig. 10 t/m 13 Regressielijnen voor NO en NO/NO_2 in verschillende perioden

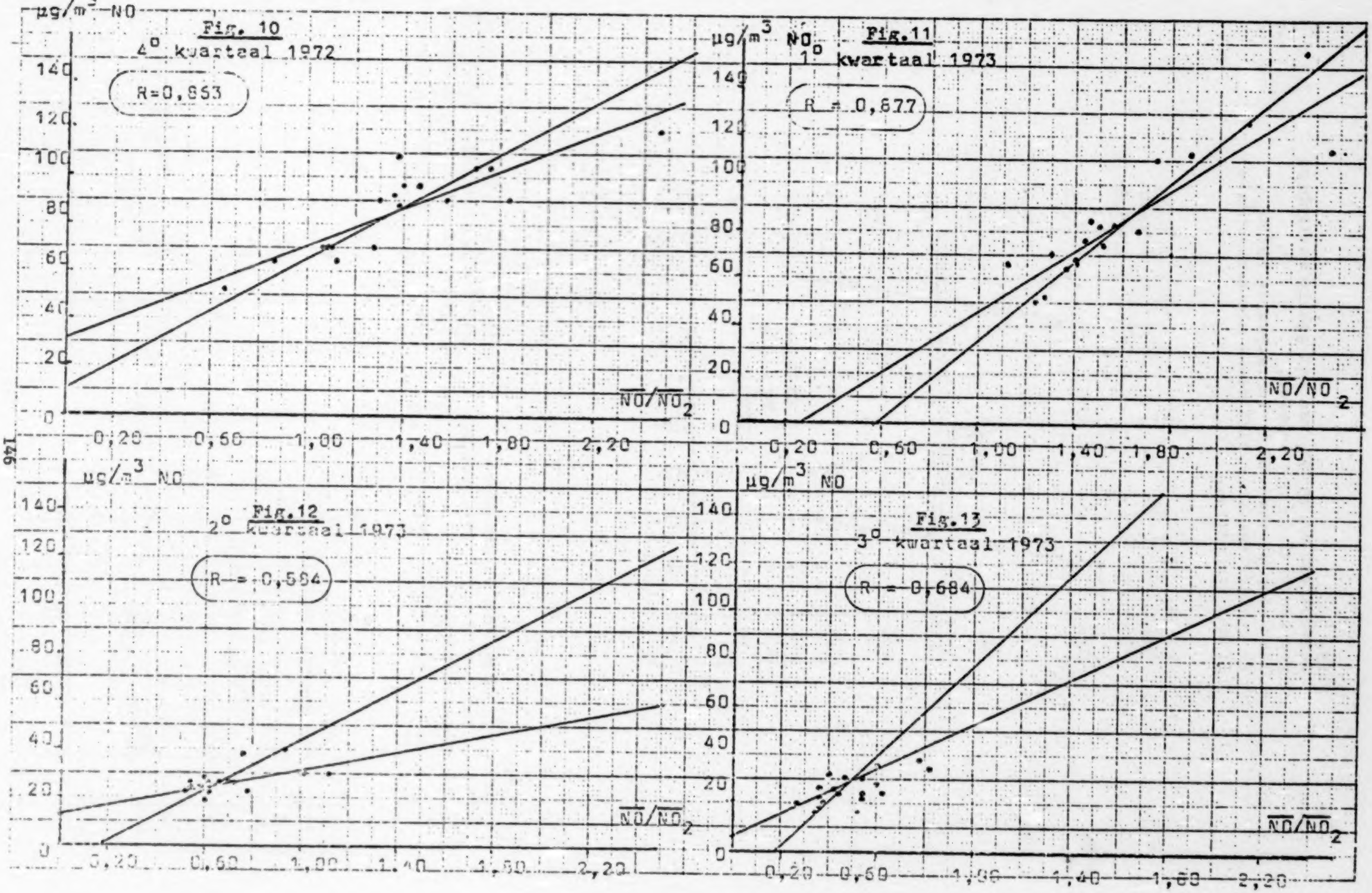


Fig.14 t/m 17 Regressielijnen voor NO en NO/NO_2 in verschillende perioden

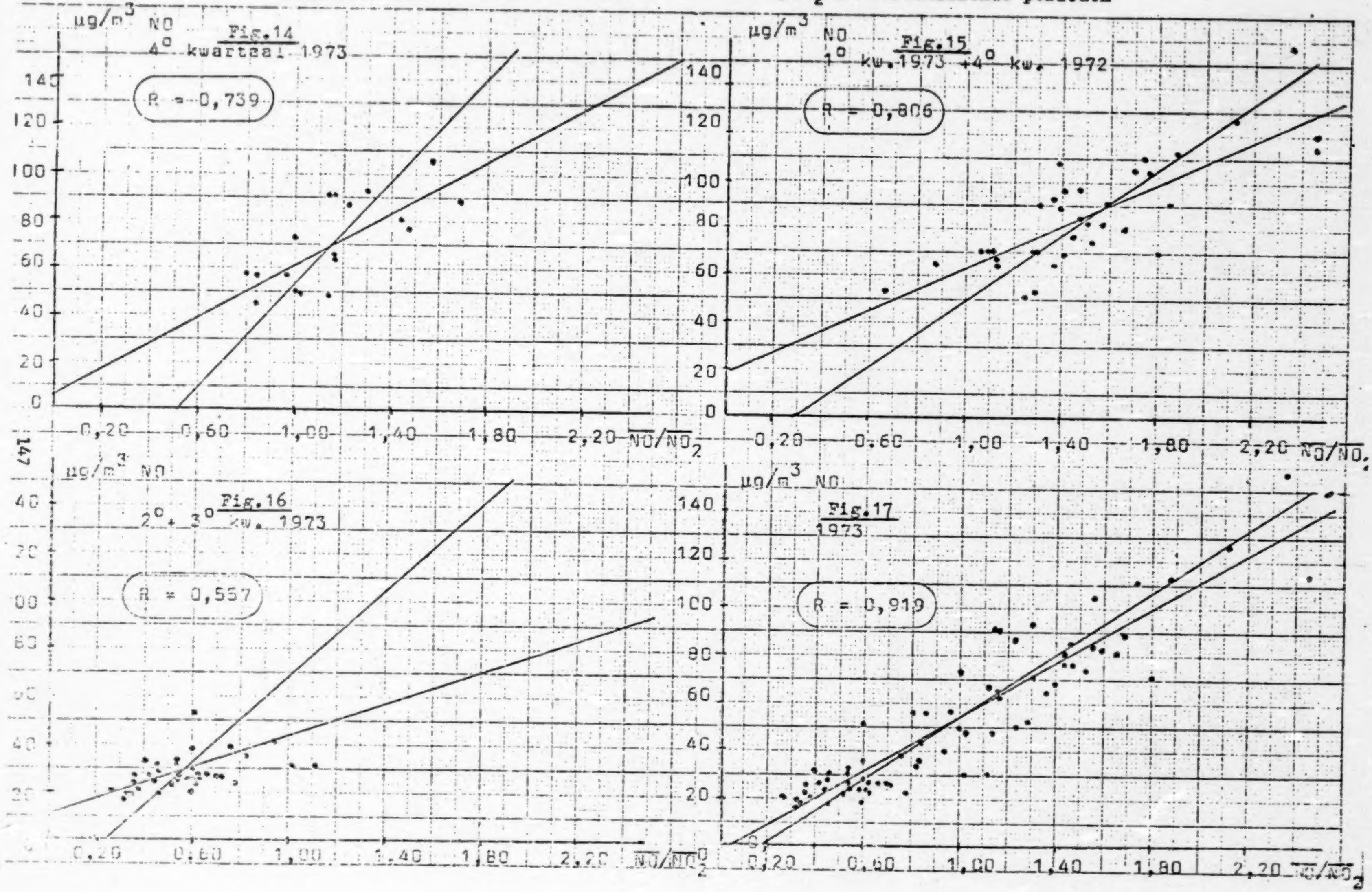
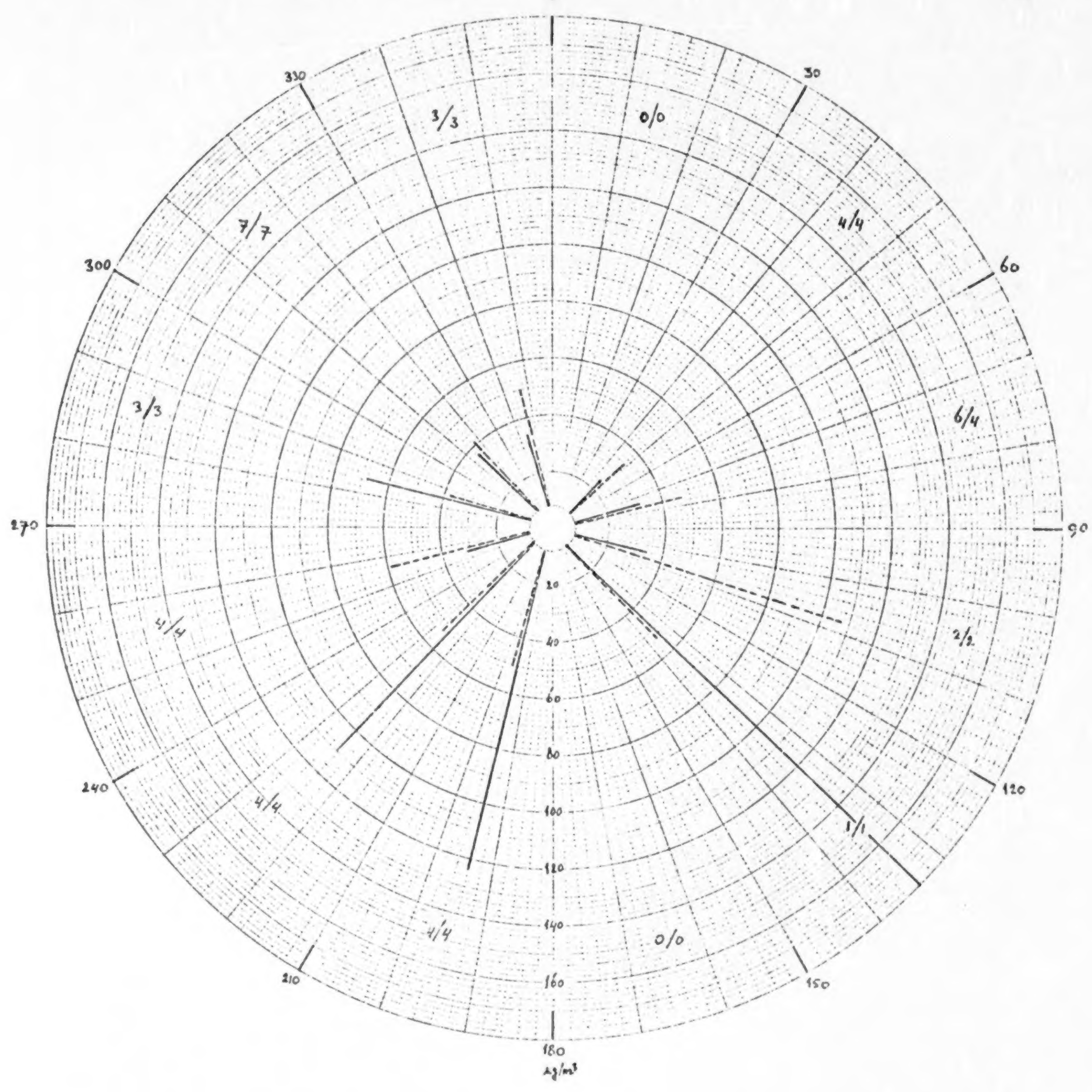


Fig. 18

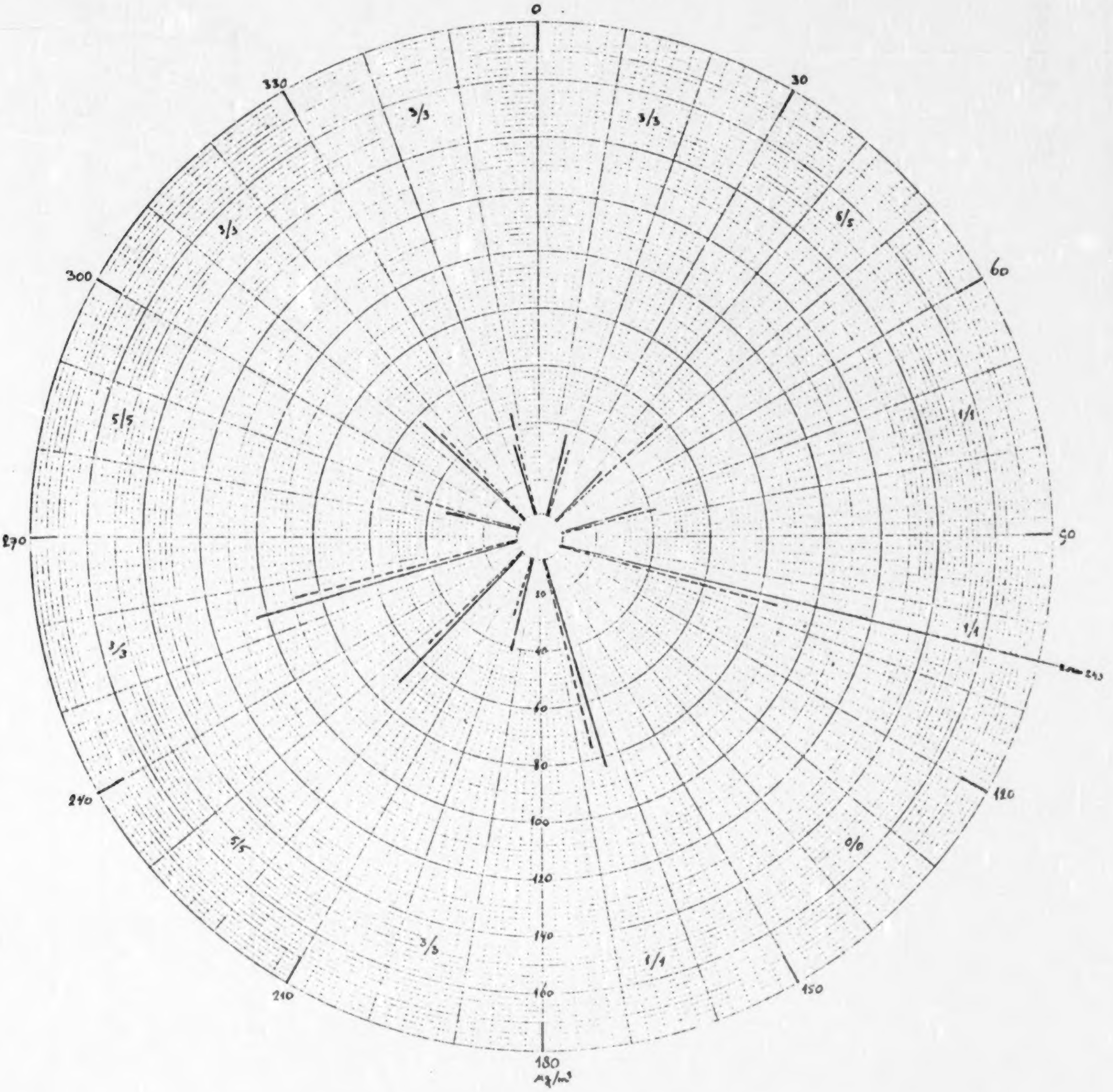
NO (——) en NO_2 (---) in 1973 op meetpunt Sweelincksingel
De gemeten waarden zijn gemiddeld per sektor van 30°.
Het aantal waarden staat in de sektor vermeld: NO/NO_2
De gemeten waarden bij variabele wind en de windsnelheid
zijn buiten beschouwing gelaten.



No. 505 H
meetpapier - wormerveer

Fig. 19

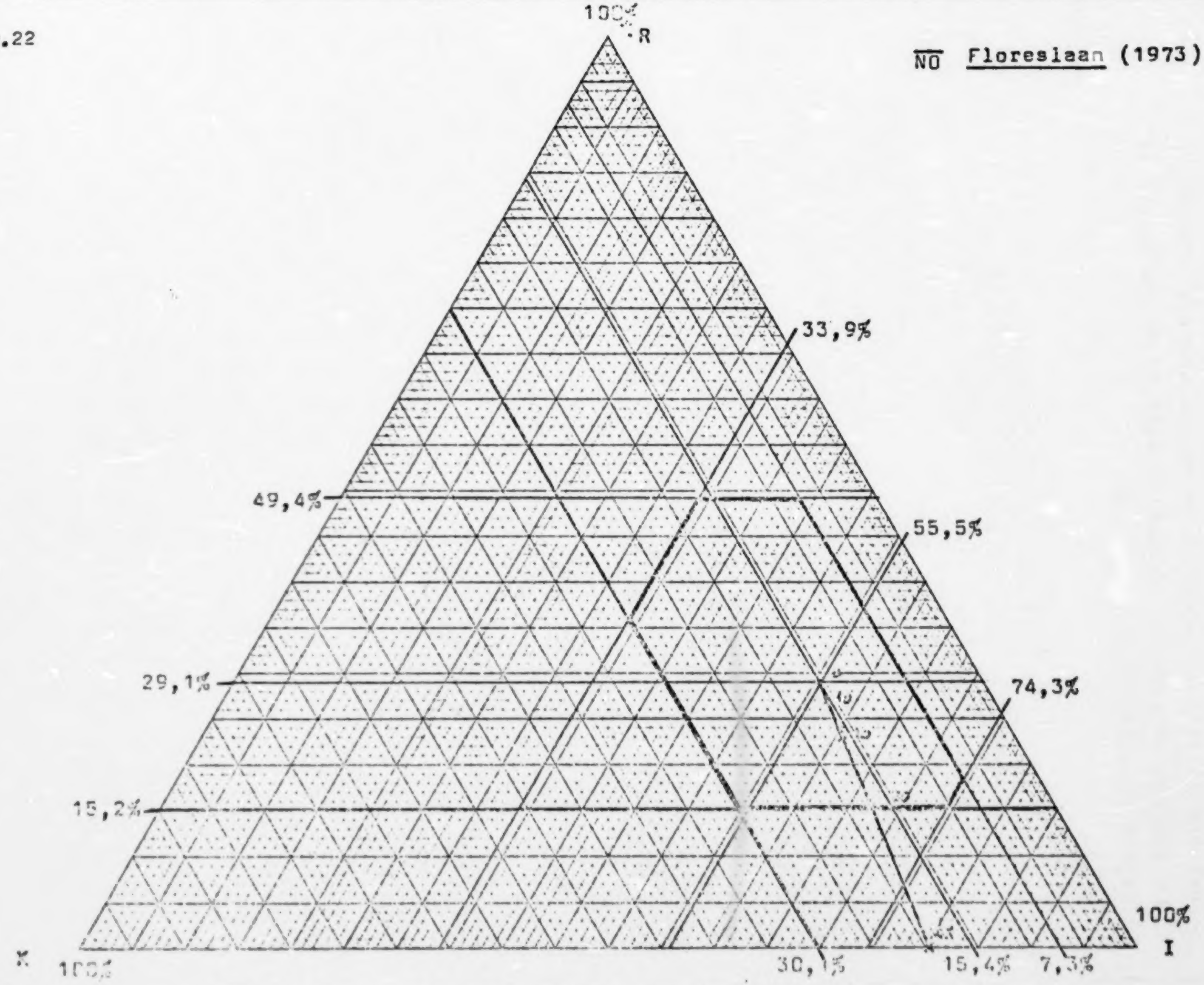
NO (——) en NO$_2$ (---) in 1973 op meetpunt Schulpweg.
De gemeten waarden zijn gemiddeld per sektor van 30^0.
Het aantal waarden staat in de sektor vermeld : NO/NO$_2$
De gemeten waarden bij variabele wind en de windsnelheid
zijn buiten beschouwing gelaten.



No. 505 H
meetpapier - wormerveer

Fig.22

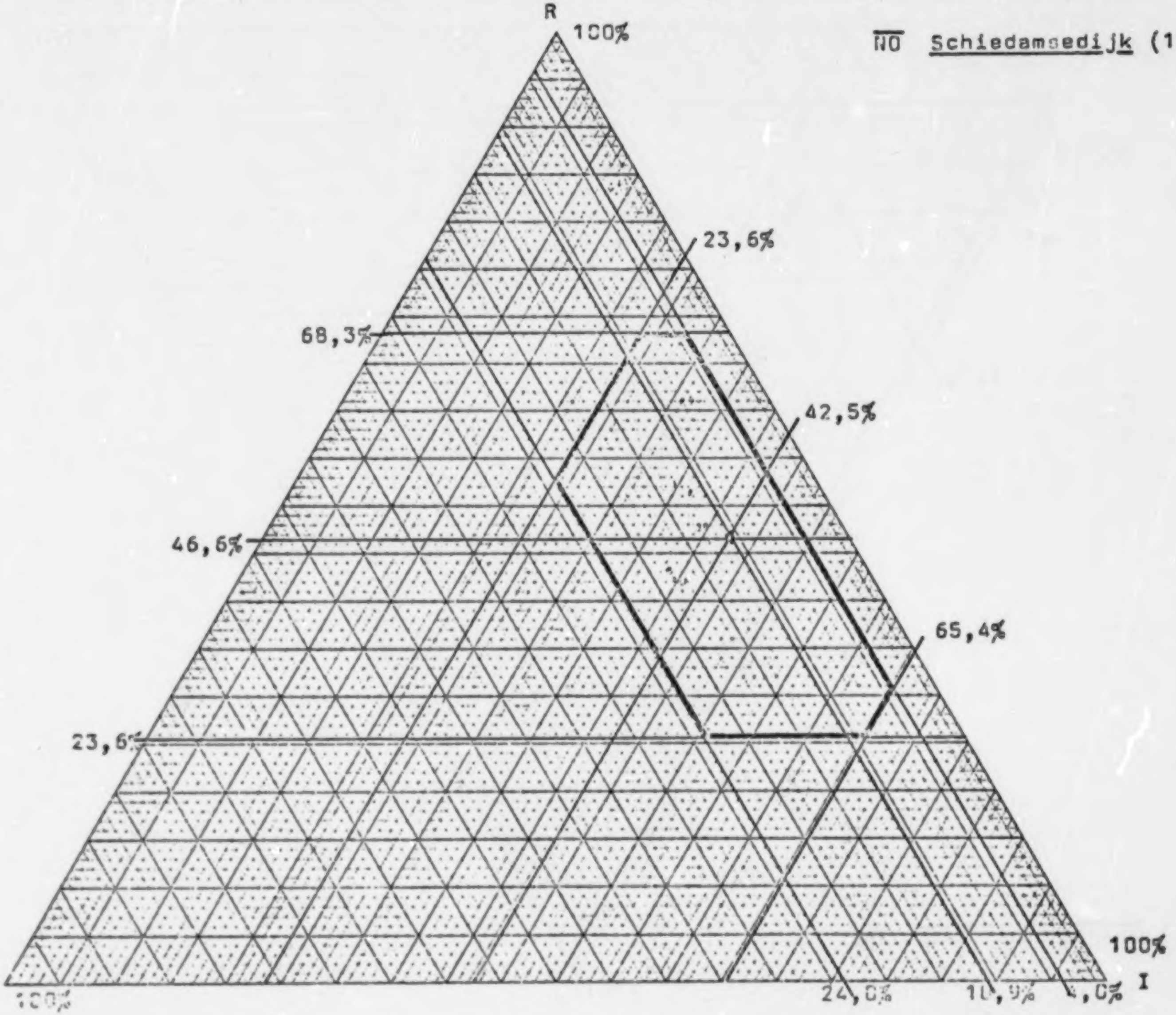
$\overline{\text{NO}}$ **Floreslaan** (1973)



No. 501TR

Fig.23

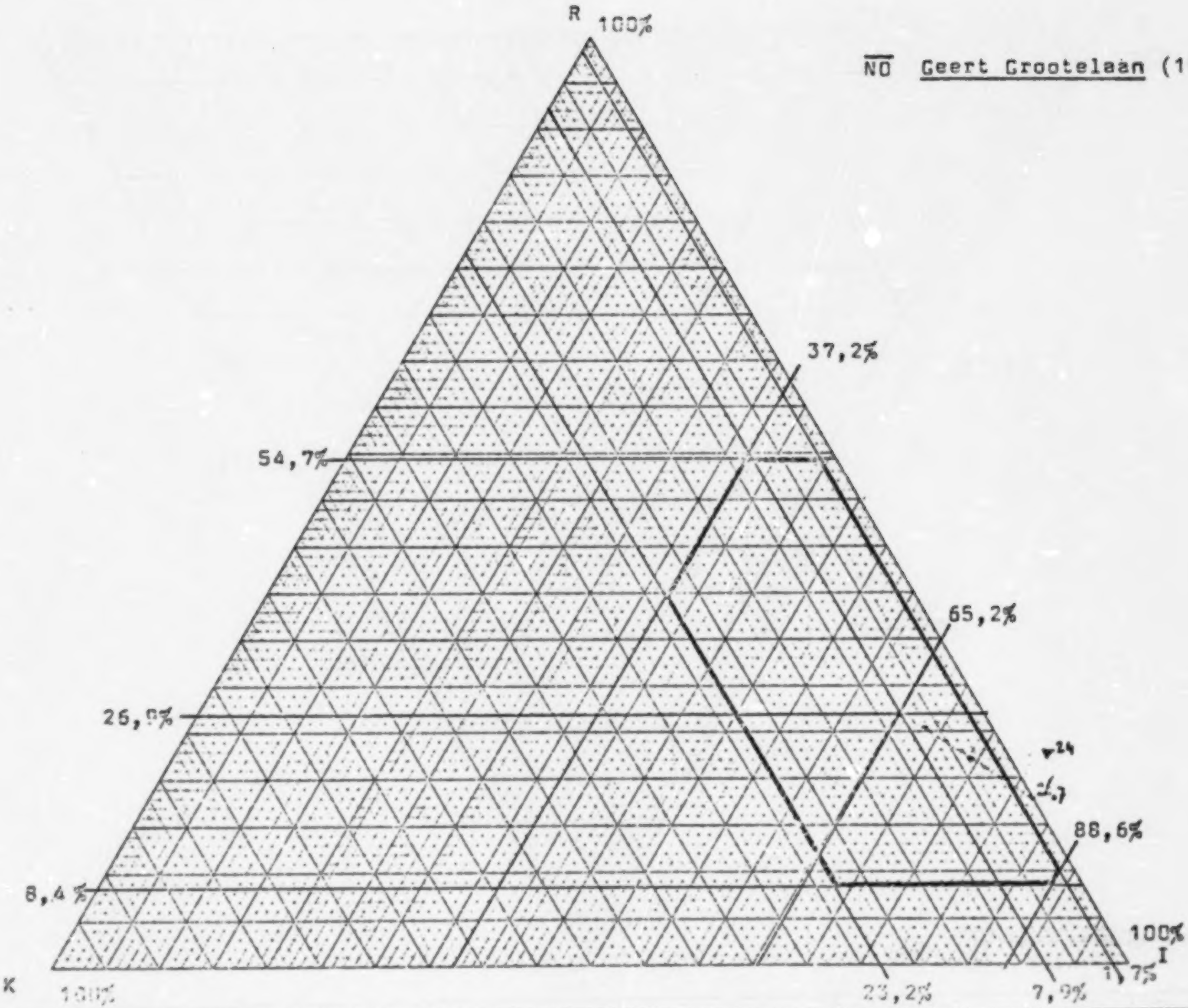
$\overline{\text{NO}}$ Schiedamsedijk (1973)



MERCURIUS - WORMERVEER
No. 501 TR

Fig.24

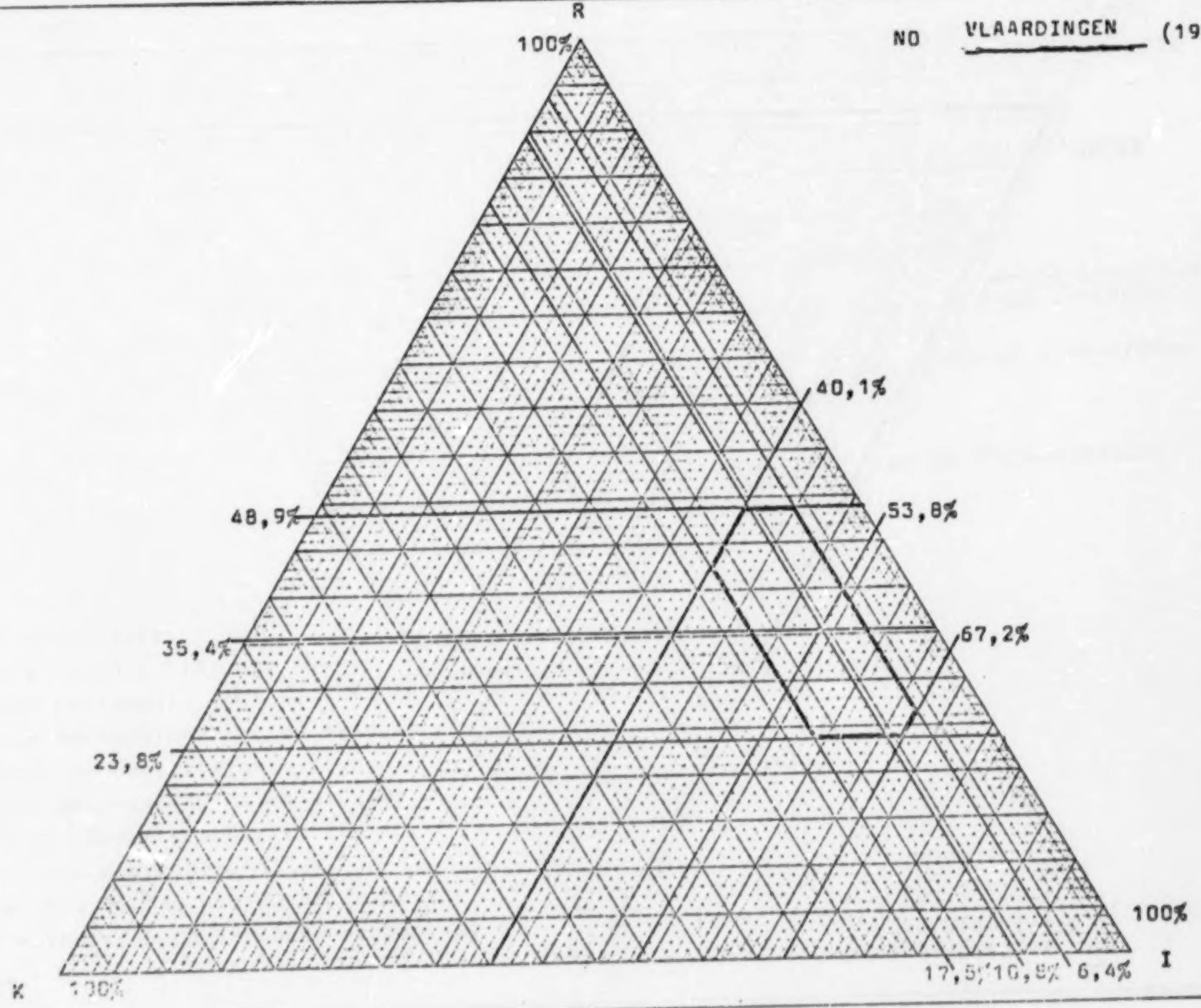
$\overline{\text{NO}}$ Geert Grootelaan (1973)



MERCURIUS-WORMERVEER
No. 501 TR

Fig.25

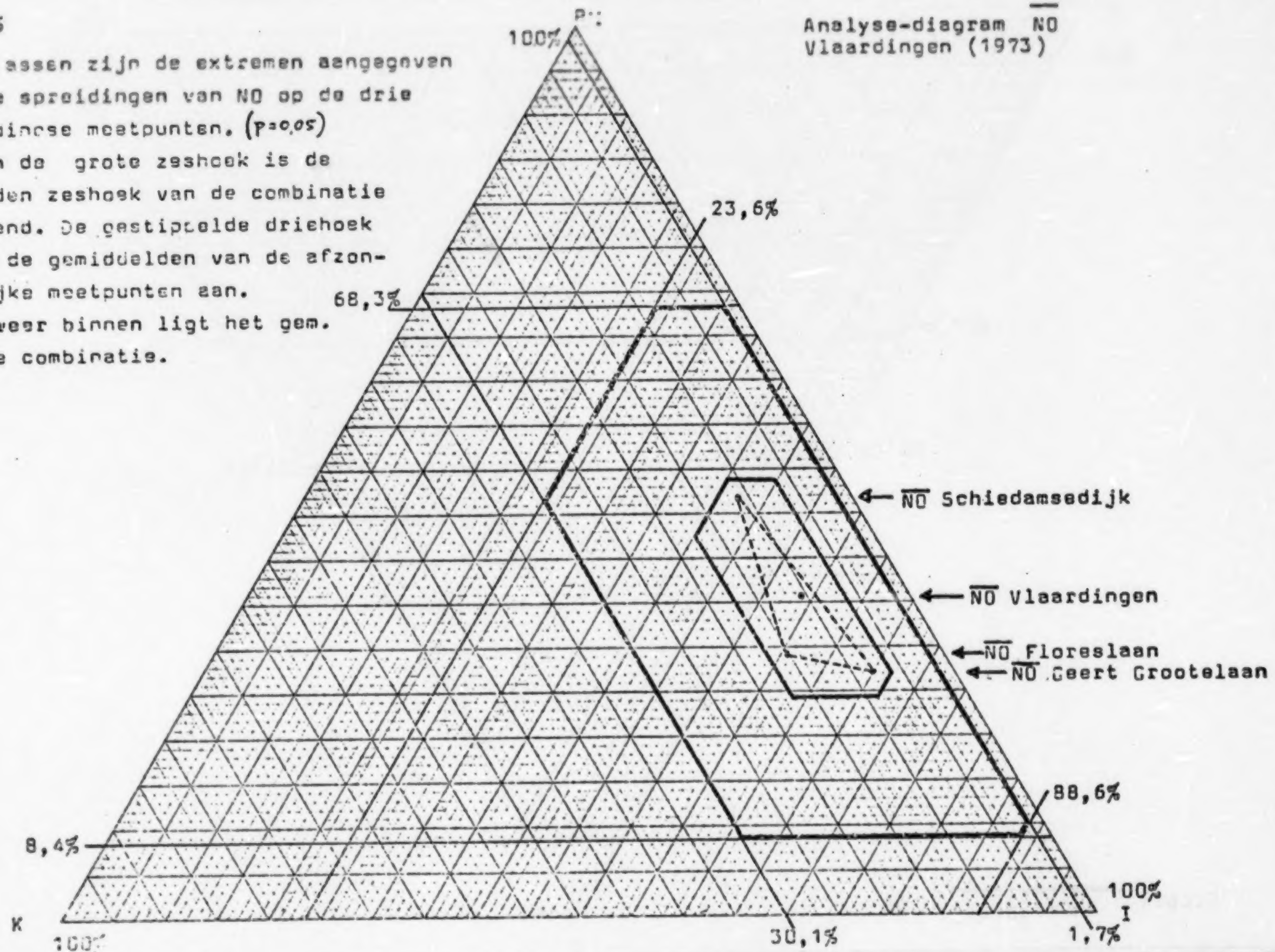
NO **VLAARDINGEN** (1973)



CURUS - WORMERVEER
No. 801 TR

Fig.26

Op de assen zijn de extremen aangegeven van de spreidingen van NO op de drie Vlaardingse meetpunten. ($p=0{,}05$)
Binnen de grote zeshoek is de gevonden zeshoek van de combinatie getekend. De gestippelde driehoek geeft de gemiddelden van de afzonderlijke meetpunten aan.
Daar weer binnen ligt het gem. van de combinatie.

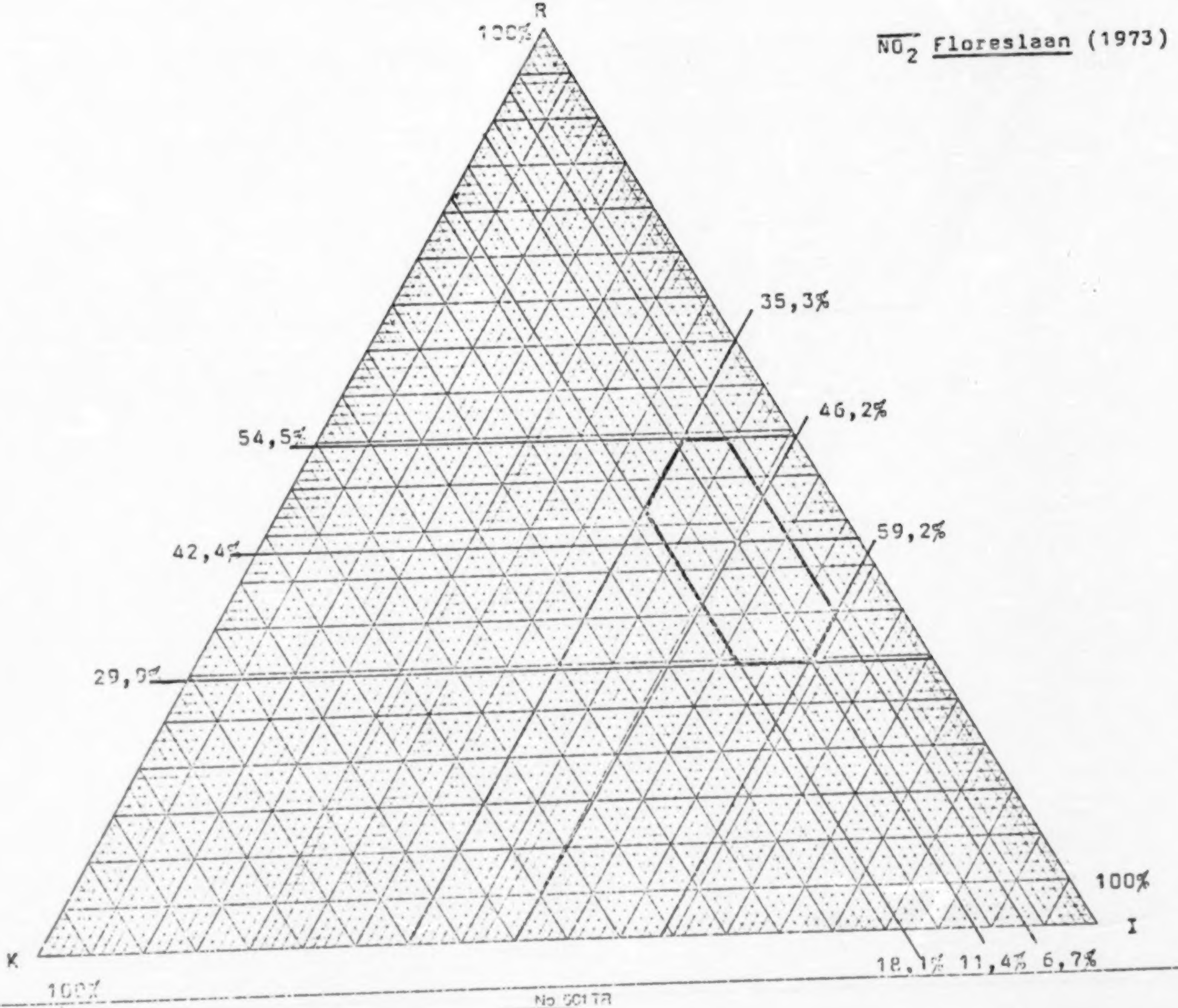


MERCURIUS - WORMERVEER

No. 501 TR

Fig.27

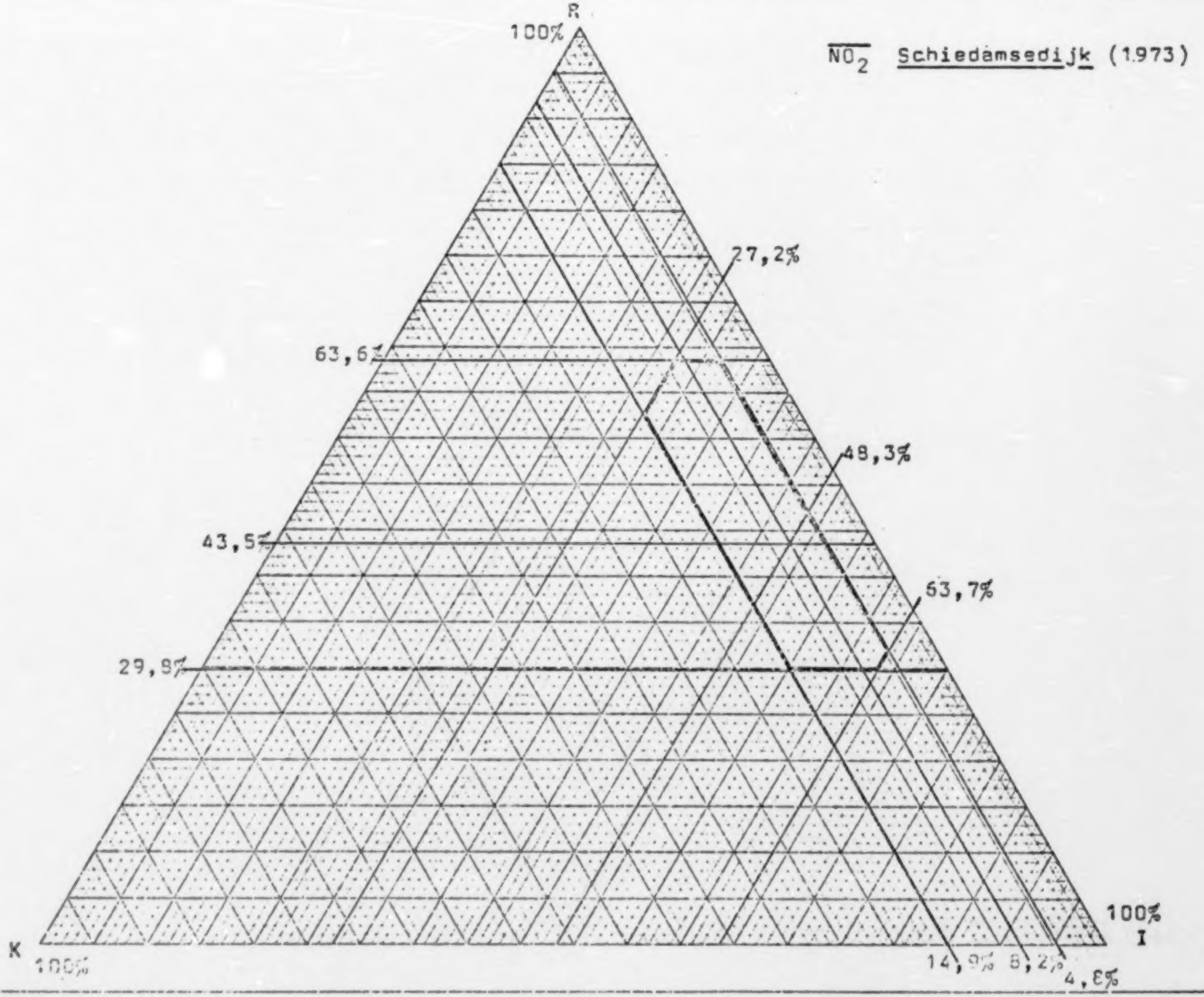
$\overline{NO_2}$ Floreslaan (1973)



AEROHAUS - WORMERVEER

Fig.28

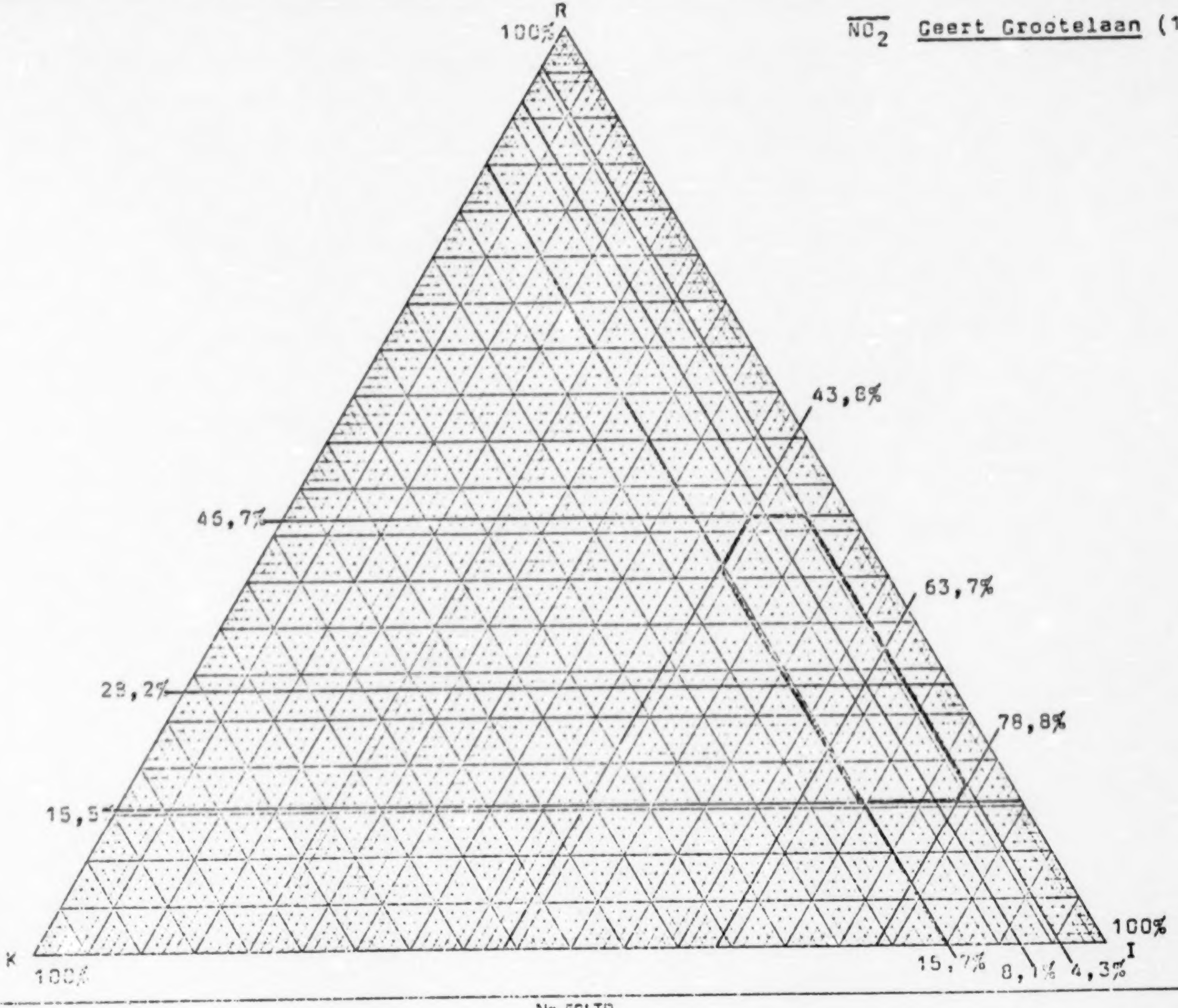
$\overline{NO_2}$ Schiedamsedijk (1973)



RCURIUS -WORMERVEER

Fig.29

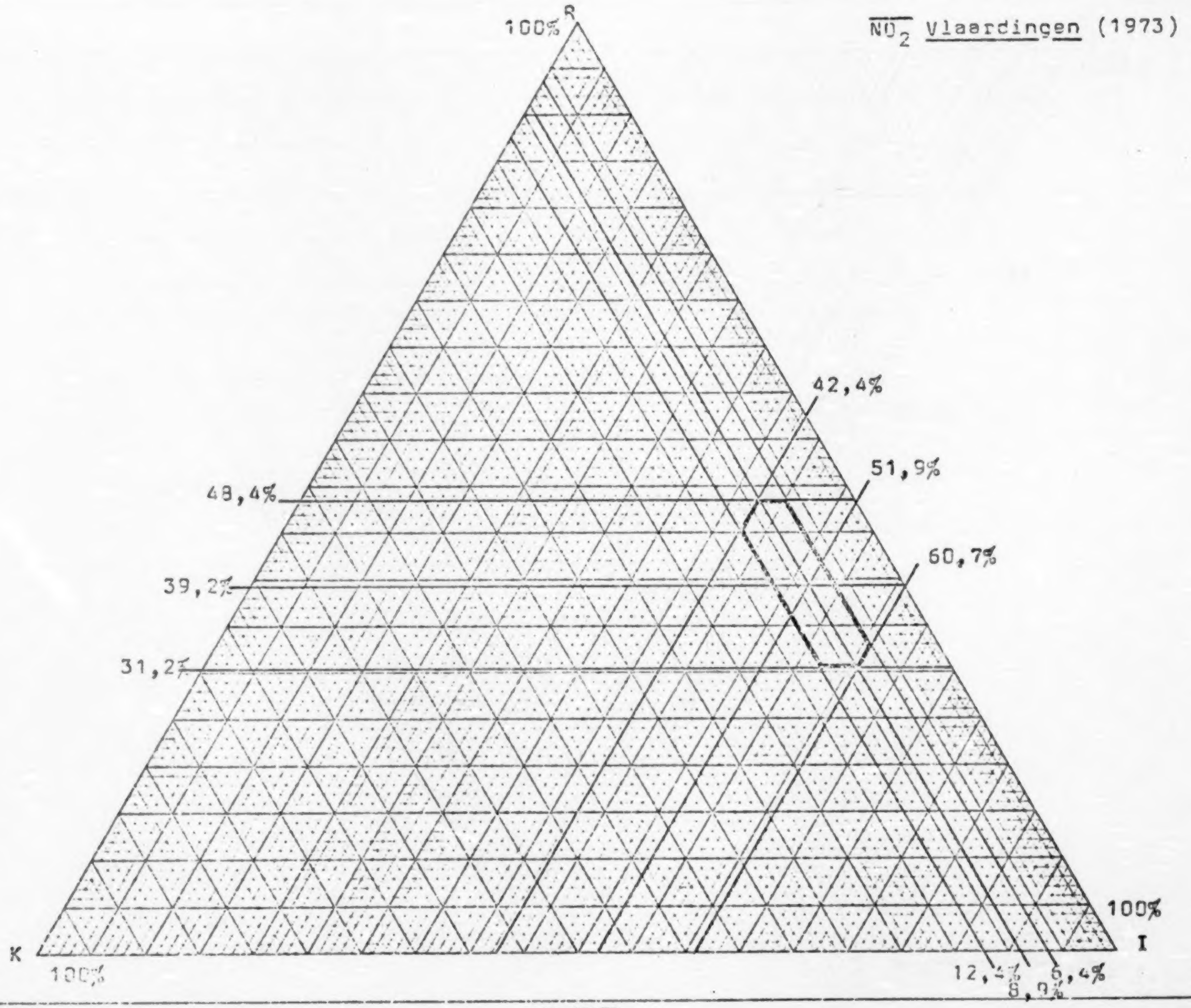
$\overline{NO_2}$ Geert Grootelaan (1973)



No. 501 TR
...CURIUS - WORMERVEER

Fig.30

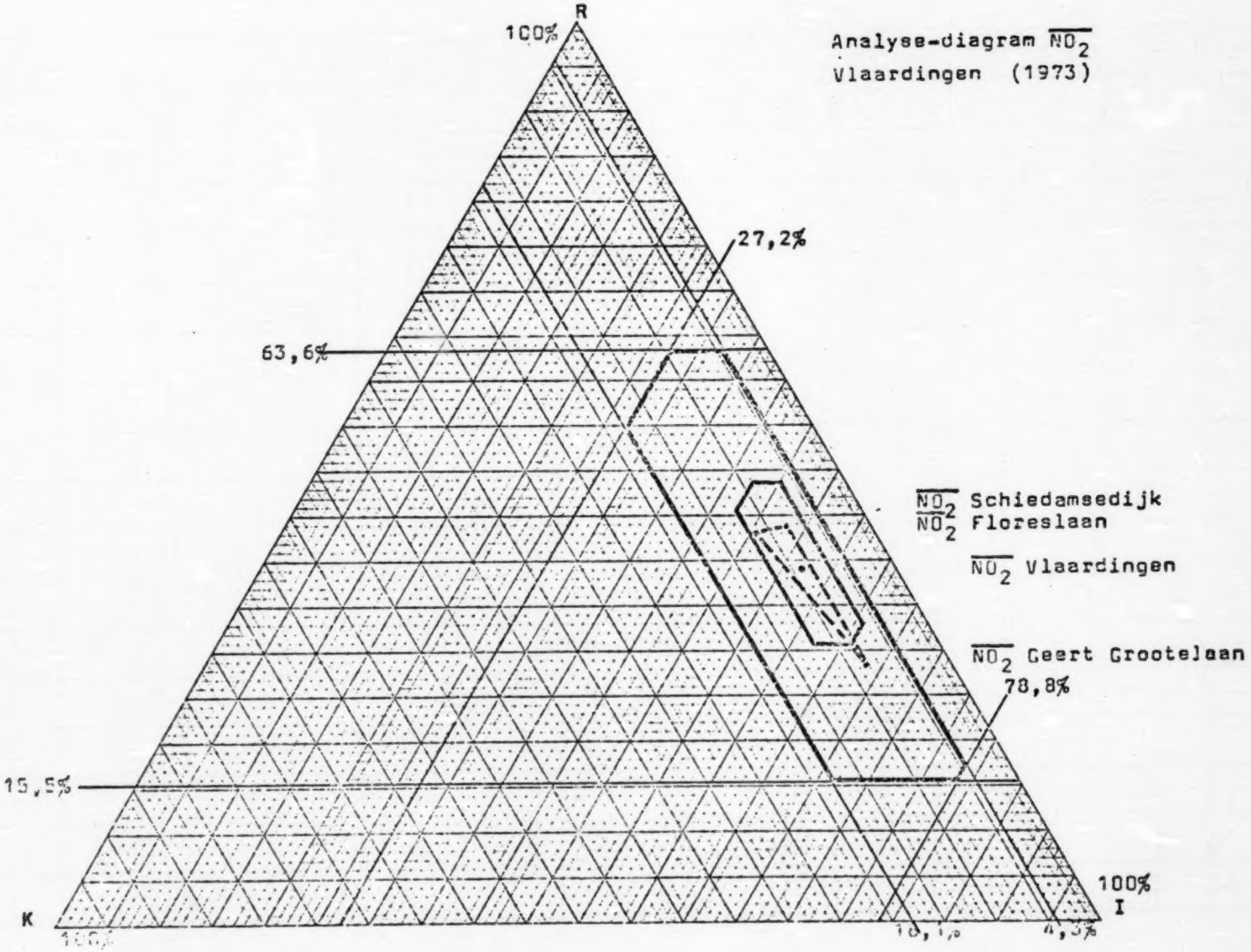
$\overline{NO_2}$ Vlaardingen (1973)



MERCURIUS - WORMERVEER

No. 50: TR

Fig.31



Analyse-diagram $\overline{NO_2}$
Vlaardingen (1973)

ROURIUS - WORMERVEER
No. 501 TR

DAGVERLOOP NO/NO$_2$ VERHOUDING

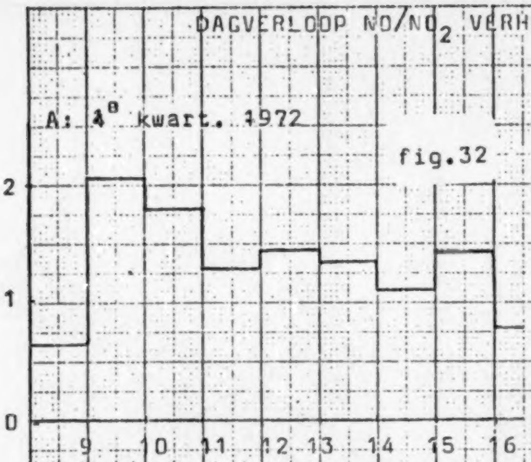
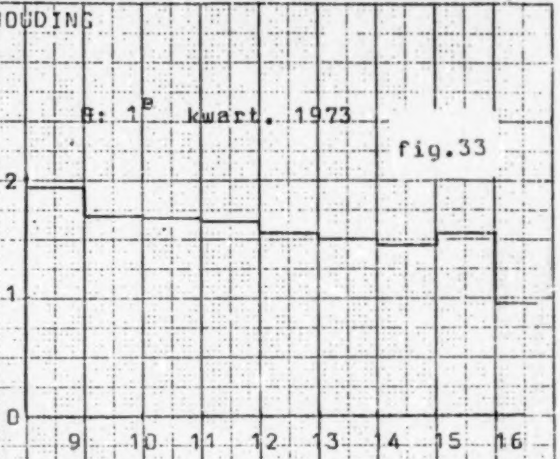


fig.32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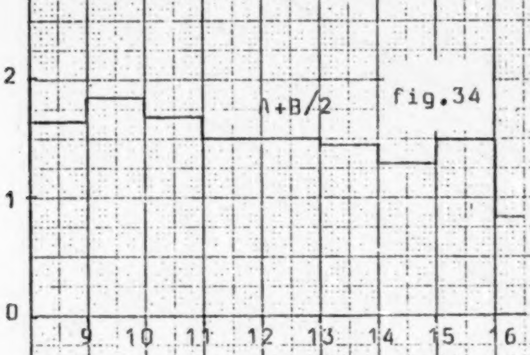


fig.34

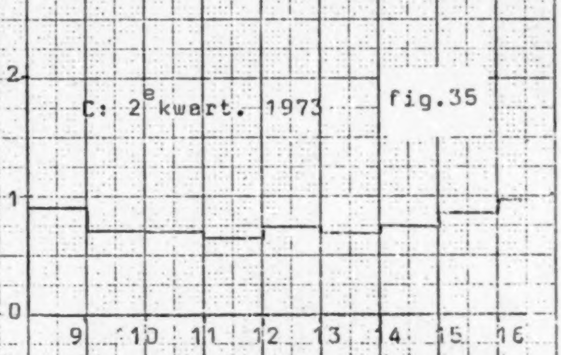


fig.35

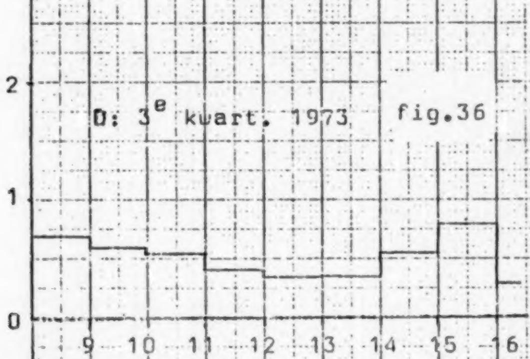
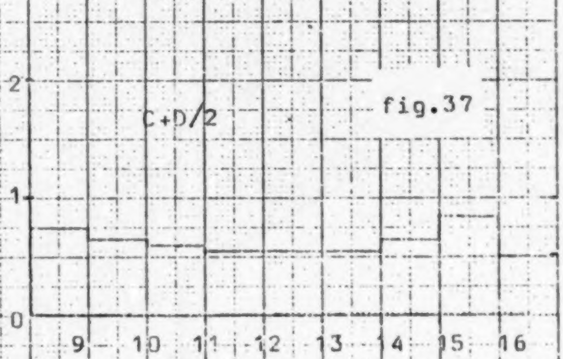


fig.36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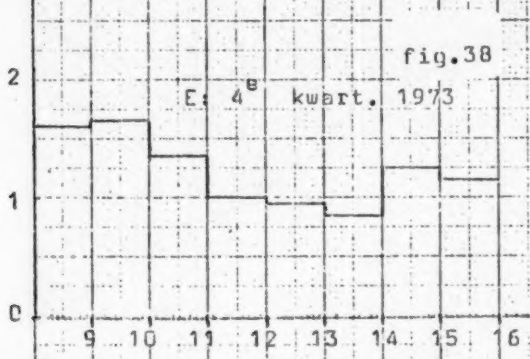


fig.38

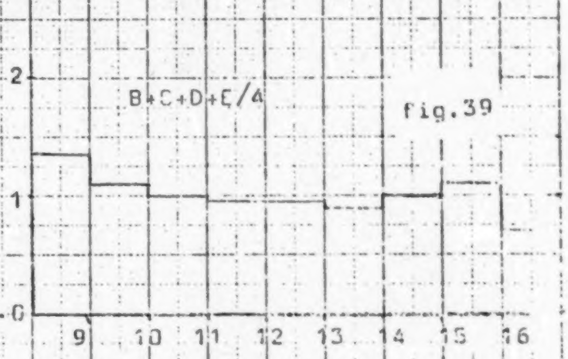


fig.39

DAGVERLOOP NO$_x$ NIVEAU

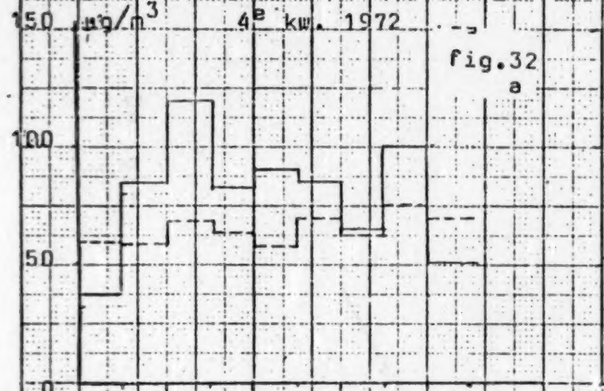


fig.32 a

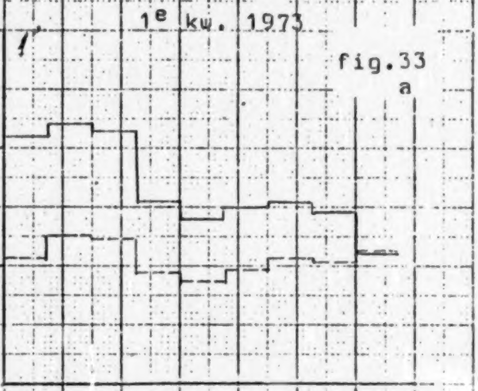


fig.33 a

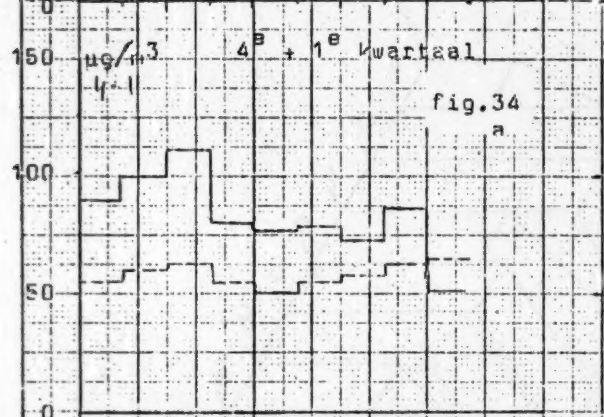


fig.34 a

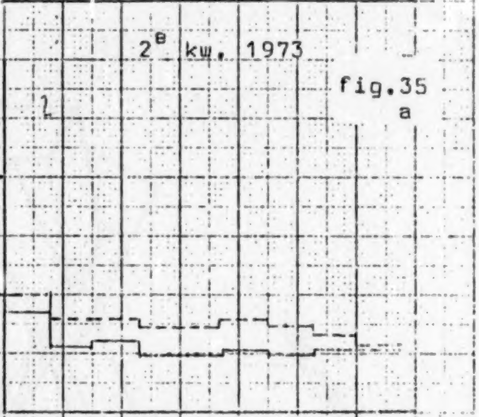
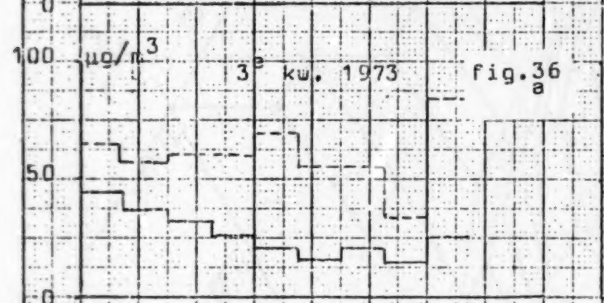


fig.35 a



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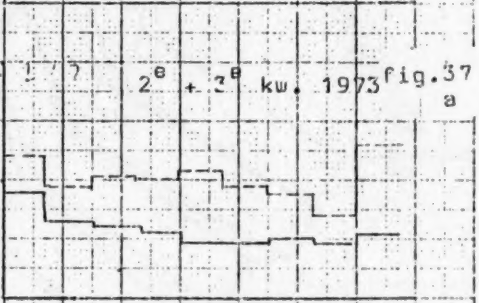


fig.37 a

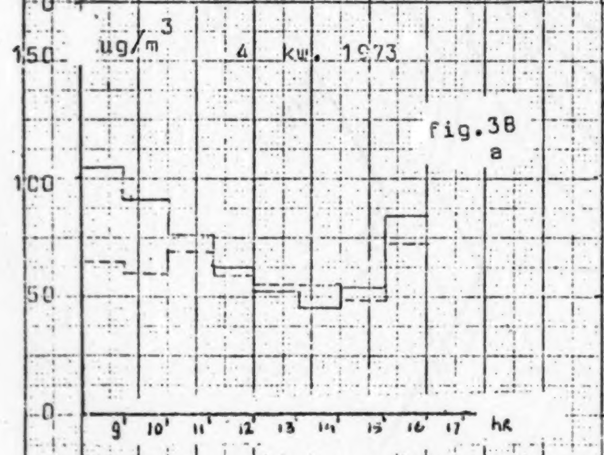
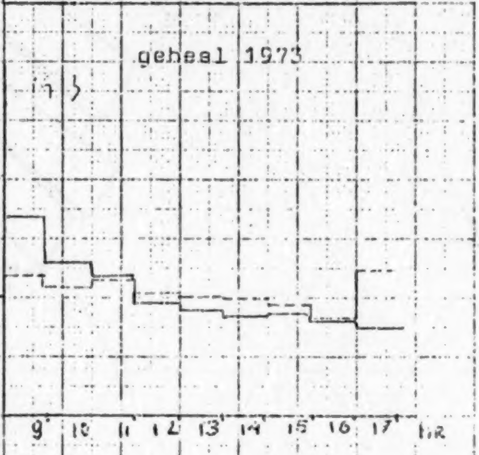


fig.38 a



(——— = NO) (– – – – =NO$_2$)

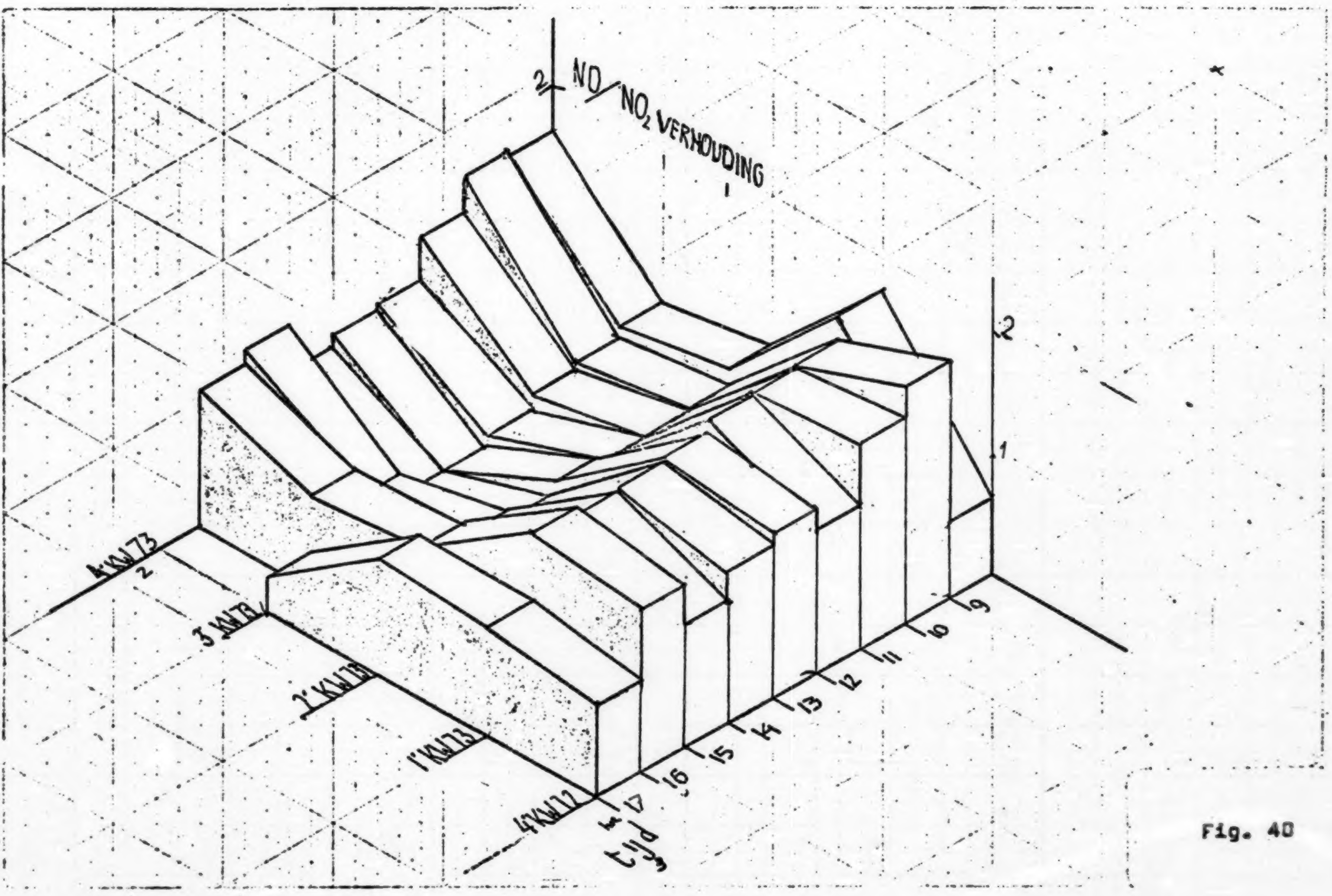
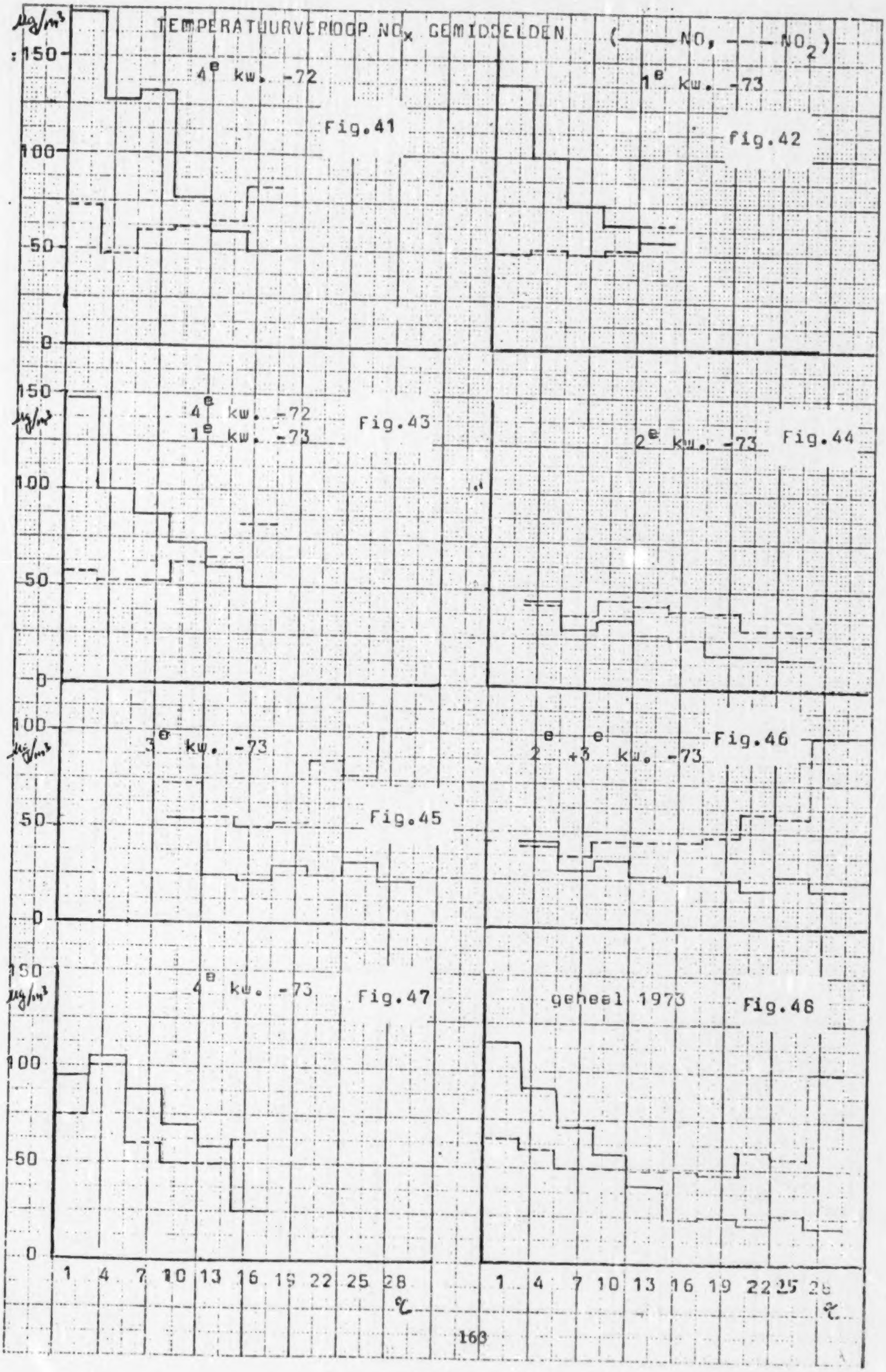

2
NO/NO$_2$ VERHOUDING
1
2
1
9
10
11
12
13
14
15
16
17
tijd
4KW73
3KW73
2KW73
1KW73
4KW72

Fig. 40

TEMPERATUURVERLOOP NO$_x$ GEMIDDELDEN (—— NO, - - - NO$_2$)



TEMPERATUURVERLOOP NO/NO2 VERHOUDING

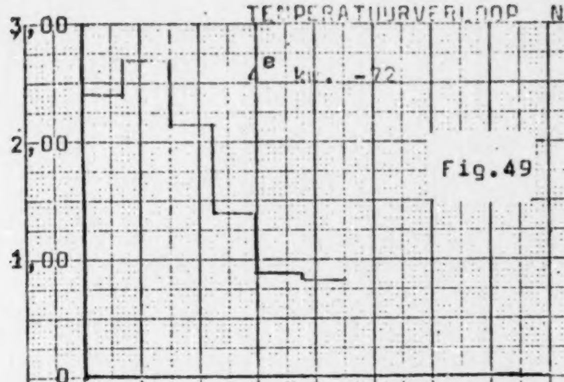


Fig.49

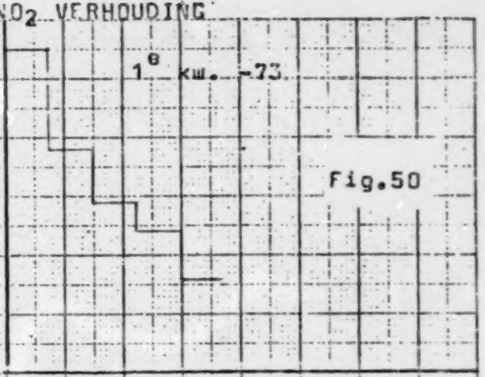


Fig.50

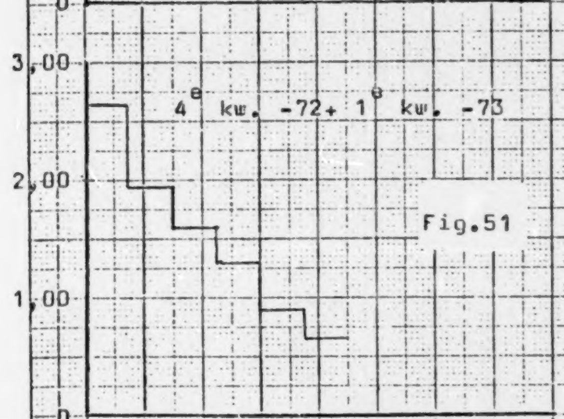


Fig.51

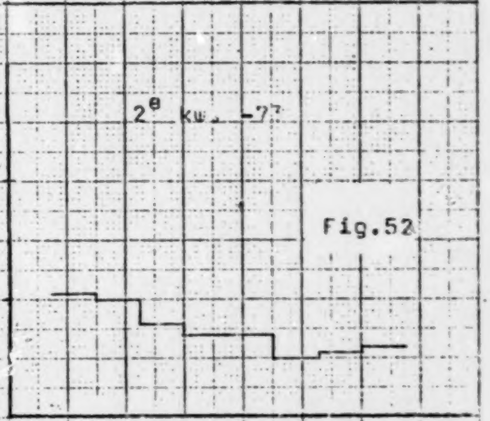


Fig.52

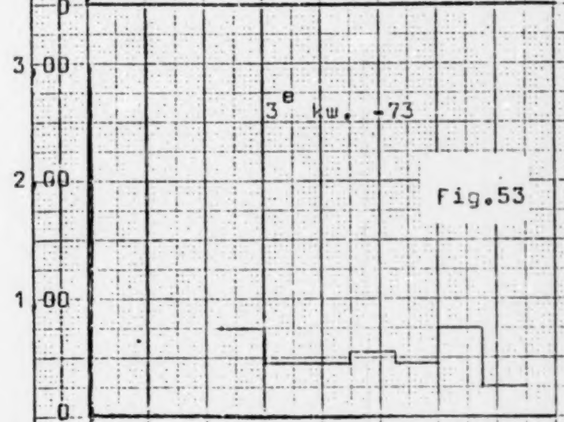


Fig.53

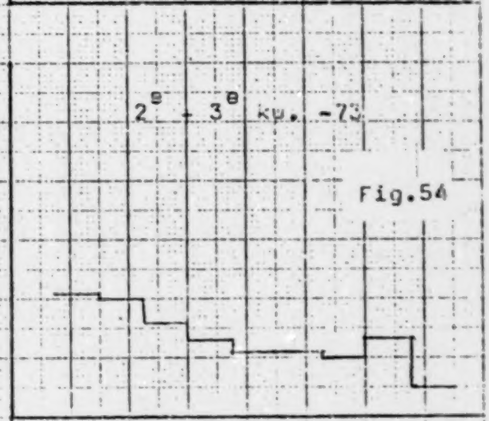


Fig.54

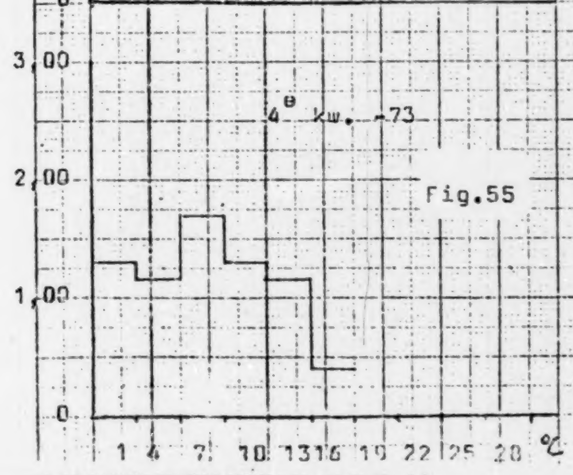


Fig.55

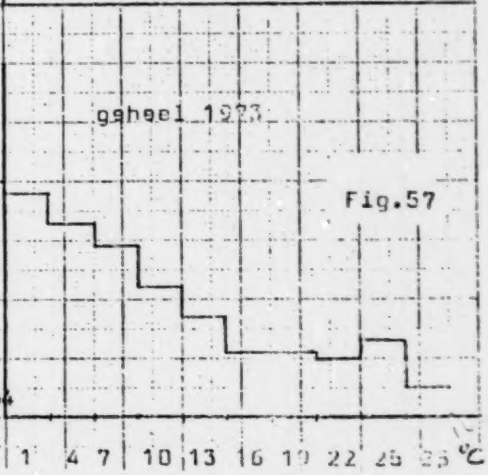


Fig.57

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